U.S. DEPARTMENT OF ENERGY
OFFICE OF CIVILIAN RADIOACTIVE WASTE MANAGEMENT

PRESENTATION TO THE NUCLEAR WASTE TECHNICAL REVIEW BOARD

SUBJECT: WASTE FORM DEGRADATION AND MATERIALS UNCERTAINTIES

PRESENTER: DR. GREGORY E. GDOWSKI

PRESENTER'S TITLE AND ORGANIZATION: CHEMICAL ENGINEER
KMI/LAWRENCE LIVERMORE NATIONAL LABORATORY
LIVERMORE, CALIFORNIA

PRESENTER'S TELEPHONE NUMBER: (510) 423-3486

OCTOBER 8 - 10, 1991

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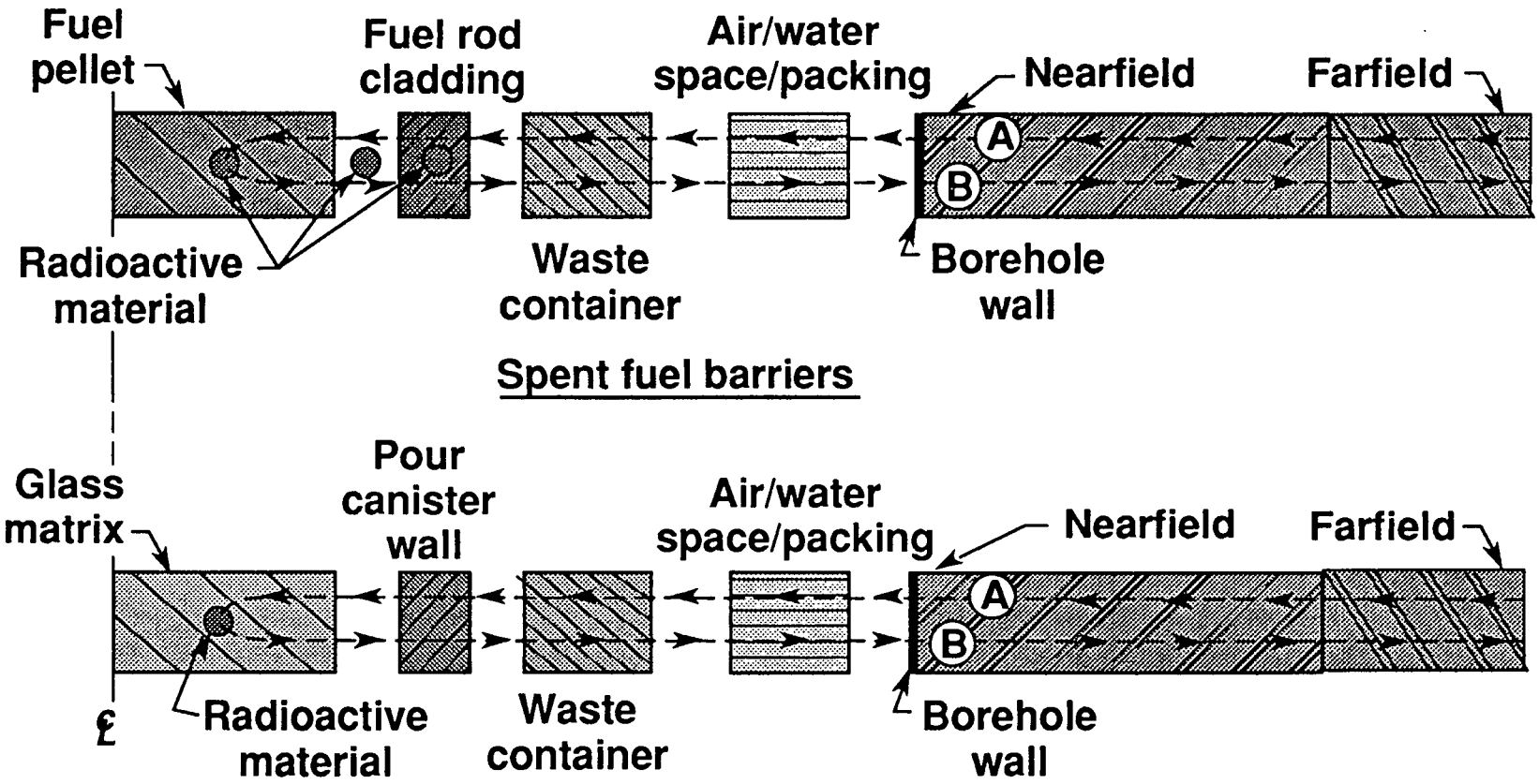
Outline of Presentation

- Introduction
- Container materials
  - Metallic alloys
- Waste form
  - Spent Fuel
    * Zircaloy cladding
    * Fuel pellets
  - Borosilicate glass
    * Pour canister
    * Glass
- Summary

Temperature Regions

- **High temperature region** well above boiling
  - Material dependent
  - Microstructural changes
  - Accelerated oxidation (corrosion)
- **Above boiling region**
  - Dominated by gas phase phenomena
  - Temperature definition is complicated by presence of hygroscopic salts, pores, and crevices
- **Below boiling region**
  - Dominated by aqueous phenomena
  - Temperature definition is complicated by presence of hygroscopic salts, pores, and crevices

Radionuclides are Isolated from the Environment by Multiple Barriers



- Failure path: Water enters from (A), radionuclides leave at (B)

Container Materials Degradation

High temperature region

- Elevated temperature (>350-500° C) phenomena
- Considerations
  - Precipitation of carbides, intermetallics
  - Graphitization
  - Internal oxidation
  - Accelerated oxidation
- Potential problems
  - All the considerations
- Potential benefits
  - None

Container Materials Degradation

(Continued)

Above boiling region

- Dry steam/air mixture with possible radiolysis products
- Considerations
  - Long-term aging
  - General corrosion (oxidation)
  - Episodic water contact
- Potential problems
  - Microstructural changes
  - Mineral deposition
  - Enhanced corrosion because of radiolysis products
- Potential benefits
  - Oxide layer growth
  - Residual stress relieving
  - Modeling

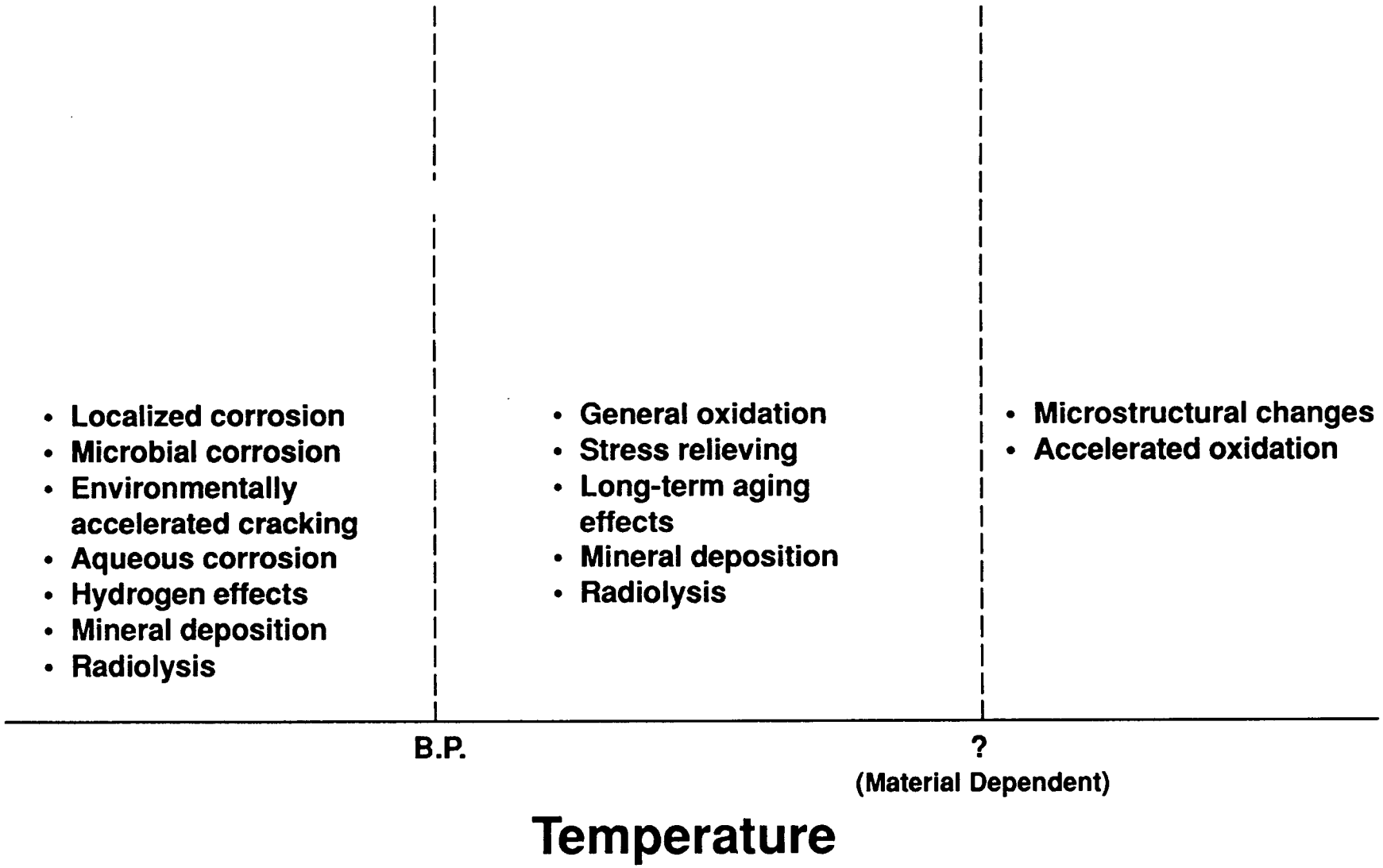
Container Materials Degradation

(Continued)

Below boiling region

- Humid air/liquid water with possible radiolysis products
- Considerations
  - General corrosion
  - Localized corrosion
  - Stress corrosion cracking
  - Microbiological corrosion
  - Hydrogen effects
  - Mineral deposition
- Potential problems
  - Corrosion processes
  - Modeling
  - Enhanced corrosion because of radiolysis products
- Potential benefits
  - Favorable water/material interaction

Temperature Regions Container Cladding



Zircaloy Cladding Degradation

High temperature region (>350° C)

- No container failure
  - Inert atmosphere
- Container failure
  - Dry steam/air mixture with possible radiolysis products
- Considerations
  - Creep/stress rupture (380° C)
  - Accelerated oxidation (540° C)
  - Internal oxidation (700° C)
- Potential problems
  - Creep/stress rupture
- Potential benefits
  - None

Zircaloy Cladding Degradation

(Continued)

Above boiling region

- **No container failure**
  - Inert atmosphere
- **Container failure**
  - Dry steam/air mixture with possible radiolysis products
- **Considerations**
  - General corrosion (oxidation)
  - Episodic water contact
  - Long-term aging
  - Radiolysis effects
- **Potential problems**
  - All the considerations
  - C-14 Release
- **Potential Benefits**
  - Above hydride precipitation temperature
  - Relieving of radiation hardening
  - Oxide layer growth
  - Modeling

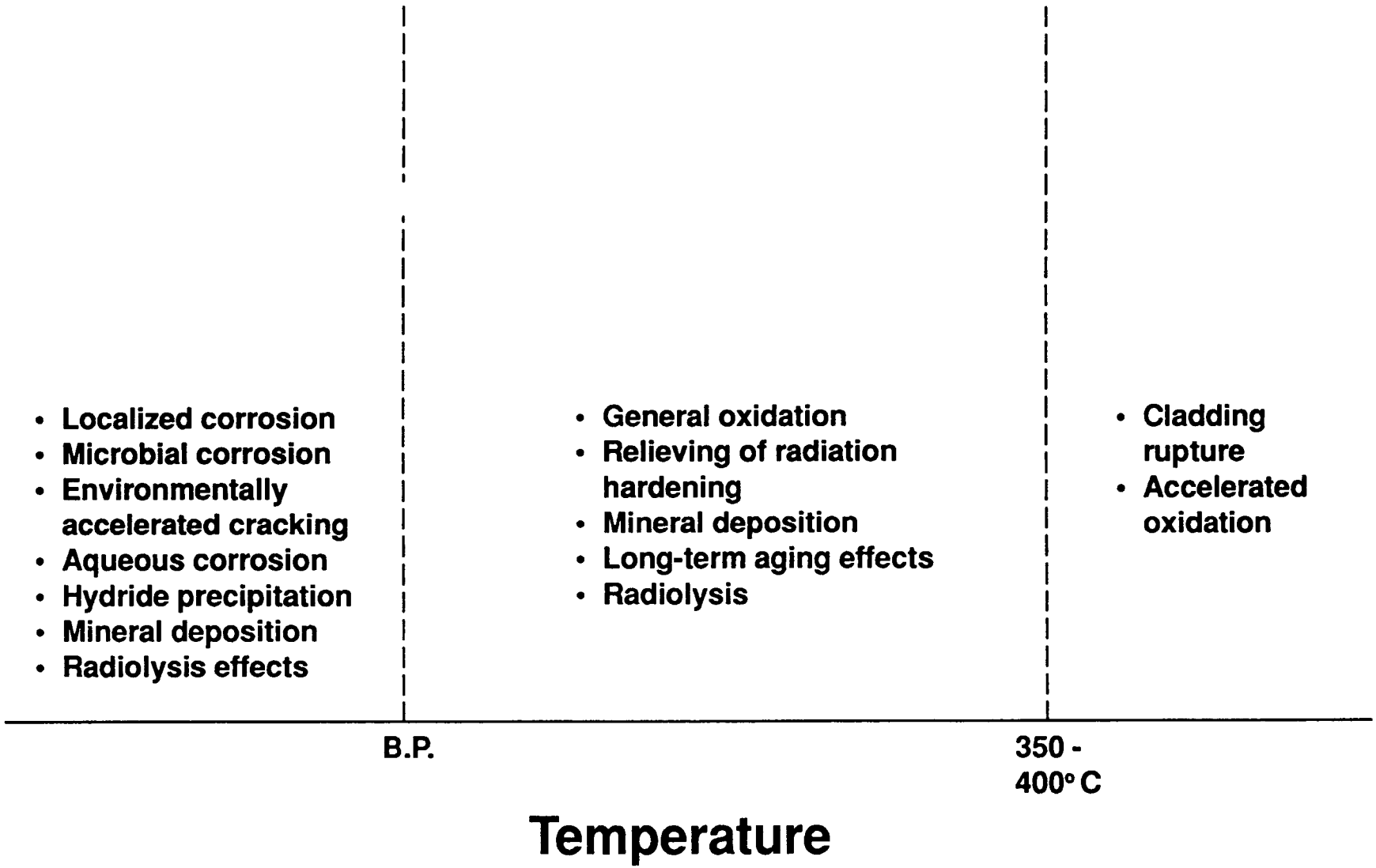
Zircaloy Cladding Degradation

(Continued)

Below boiling region

- **No container failure**
  - Inert atmosphere
- **Container failure**
  - Humid air/liquid water with possible radiolysis products
- **Considerations**
  - Localized corrosion
  - General corrosion
  - Stress corrosion cracking
  - Hydrogen effects
  - Microbiological corrosion
  - Mineral deposition
- **Potential problems**
  - All the considerations
  - Modeling
- **Potential benefits**
  - Favorable water/Zircaloy interaction

Temperature Regions Zircaloy Cladding



Fuel Pellet Degradation

Above boiling and high temperature regions

- **No container/cladding failure**
  - Inert atmosphere
- **Container/cladding failure**
  - Dry steam/air mixture with possible radiolysis products
- **Considerations**
  - Oxidation response

    >250° C U_3O_8/UO_3 (powder)

    <250° C $UO_{2.4}$ (fragments intact)
- **Potential problems**
  - Oxidation of fuel pellets and release of volatile radionuclides
- **Potential benefits**
  - No dissolution
  - No oxidation if no container/cladding failure

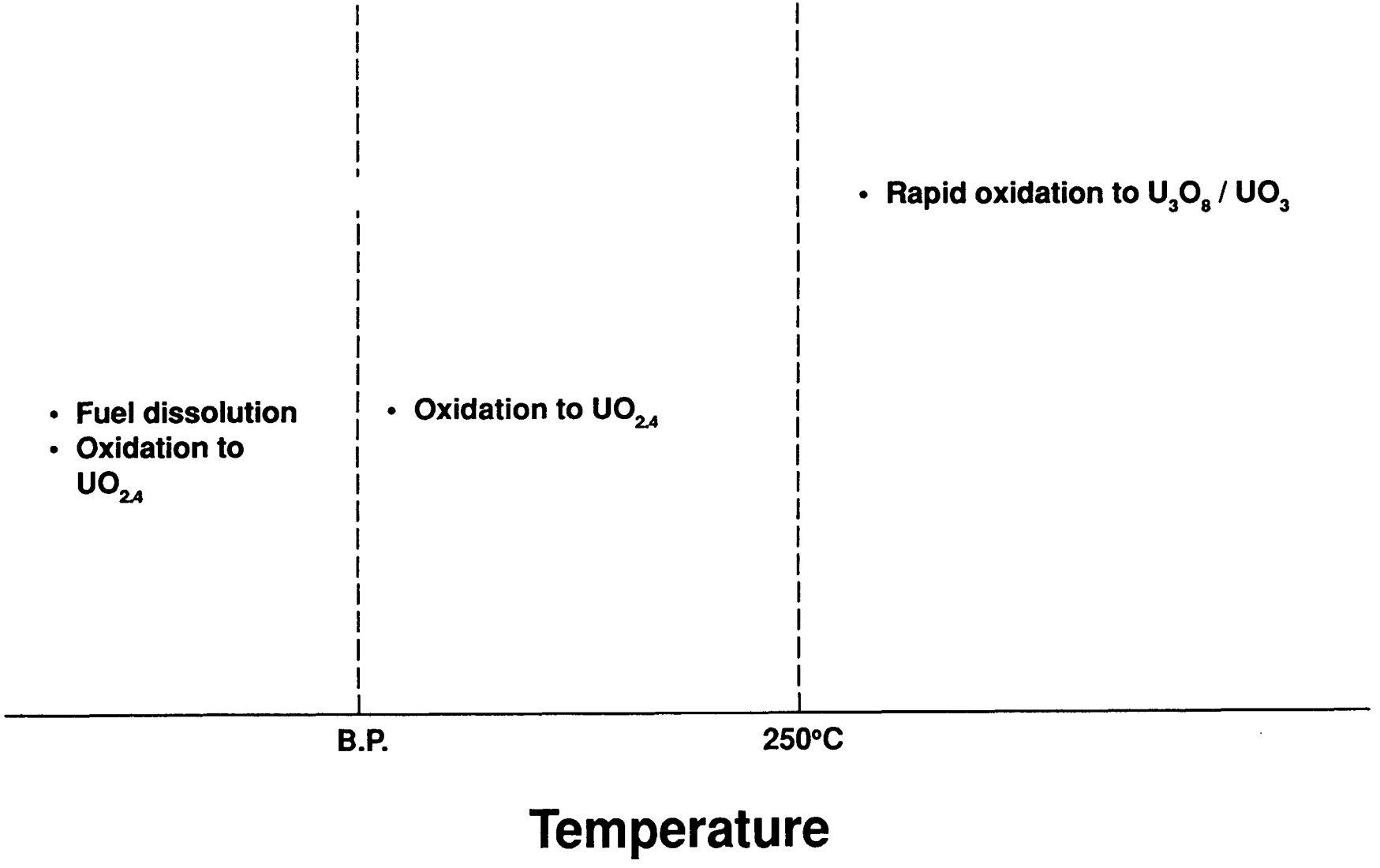
Fuel Pellet Degradation

(Continued)

Below boiling region

- **No container/cladding failure**
  - Inert atmosphere
- **Container/cladding failure**
  - Humid air/liquid water with possible radiolysis products
- **Considerations**
  - Oxidation response
  - Fuel dissolution
- **Potential problems**
  - Fuel dissolution
    - * UO_2 fragment dissolution
    - * U_3O_8/UO_3 powder dissolution
- **Potential benefits**
  - Favorable water/fuel pellet interaction
  - Low oxidation rates
  - No oxidation/dissolution if no container/cladding failure

Temperature Regions UO_2 Fuel Pellets



Borosilicate Glass Degradation

Above boiling and high temperature region

- **No container/canister failure**
  - Inert atmosphere
- **Container/canister failure**
  - Dry steam/air mixture with possible radiolysis products
- **Considerations**
  - Devitrification above 500-600° C
  - Hydration of glass
- **Potential probems**
  - Hydration of glass
- **Potential benefits**
  - Hydration rates low in low relative humidity
  - No dissolution
  - Secondary mineral precipitation
  - No hydration if no container/canister failure

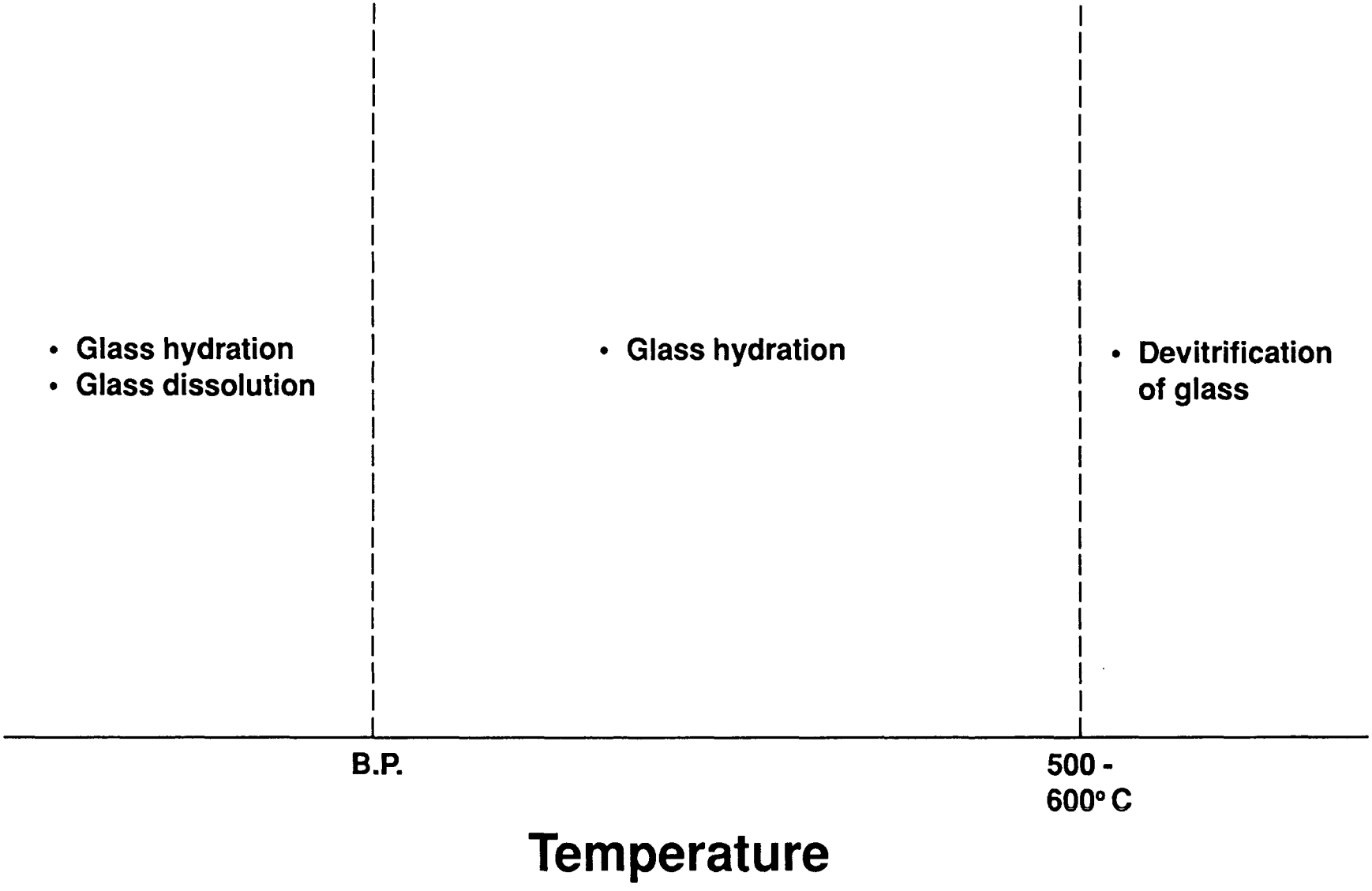
Borosilicate Glass Degradation

(Continued)

Below boiling region

- No container/canister failure
  - Inert atmosphere
- Container/canister failure
  - Humid air/liquid water with possible radiolysis products
- Considerations
  - Glass dissolution
  - Hydration of glass
- Potential problems
  - Glass dissolution
  - Hydration of glass
- Potential benefits
  - Slow hydration rates
  - Favorable water/glass interaction

Temperature Regions Borosilicate Glass



Summary

- Based on previous experience and preliminary YMP testing certain temperature regions appear to offer advantages over other temperature regions for various waste package components when considered independently:

Container materials	above boiling
Zircaloy cladding	above boiling
UO_2 fuel pellets	below boiling
Borosilicate glass	below boiling

- Testing will be necessary to determine whether degradation modes exist under repository relevant conditions, and if they exist to determine their significance

argues for robust container