Depleted Uranium Dioxide (DUO_2) as an SNF Waste-Package Particulate Fill: Fill Behavior

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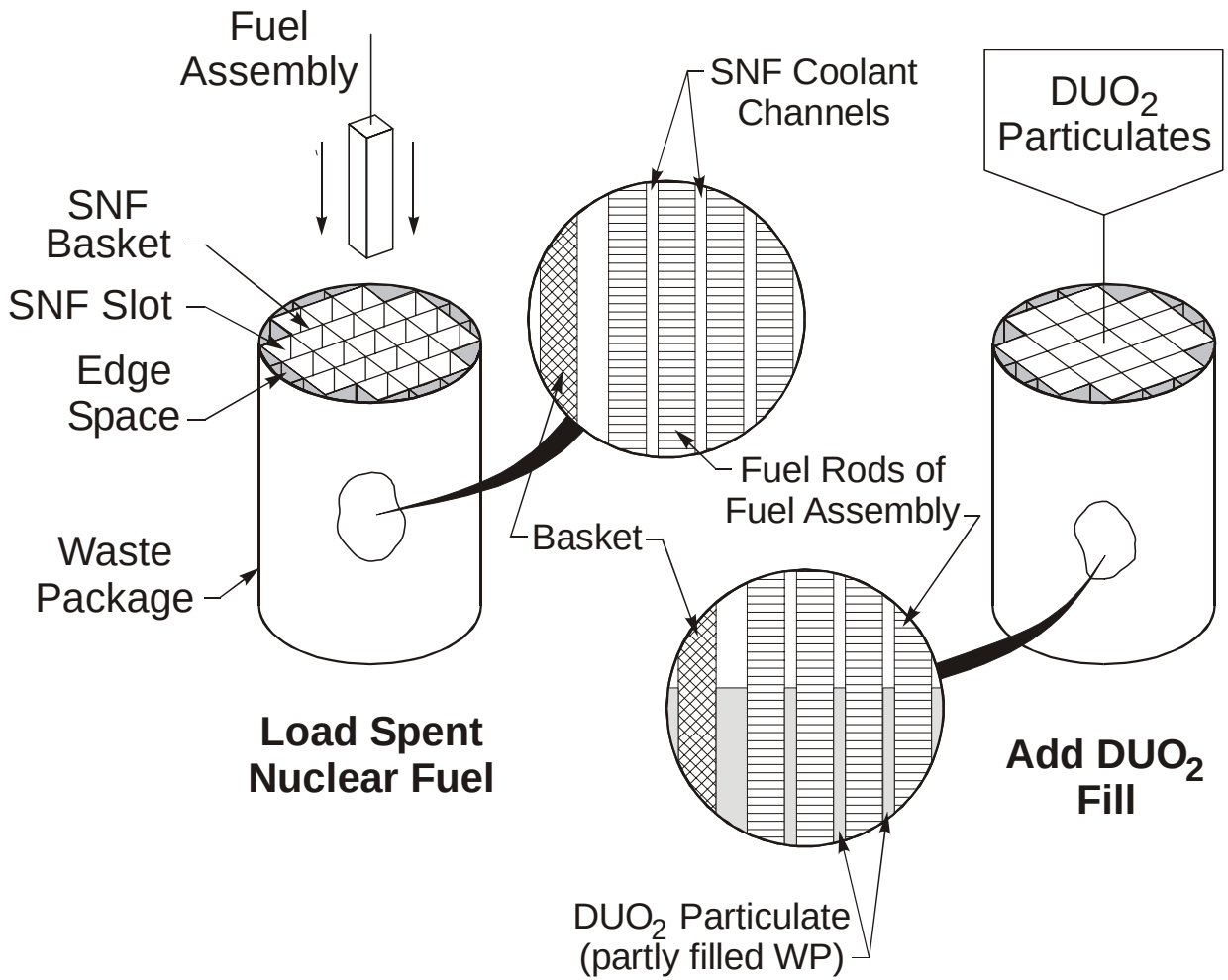
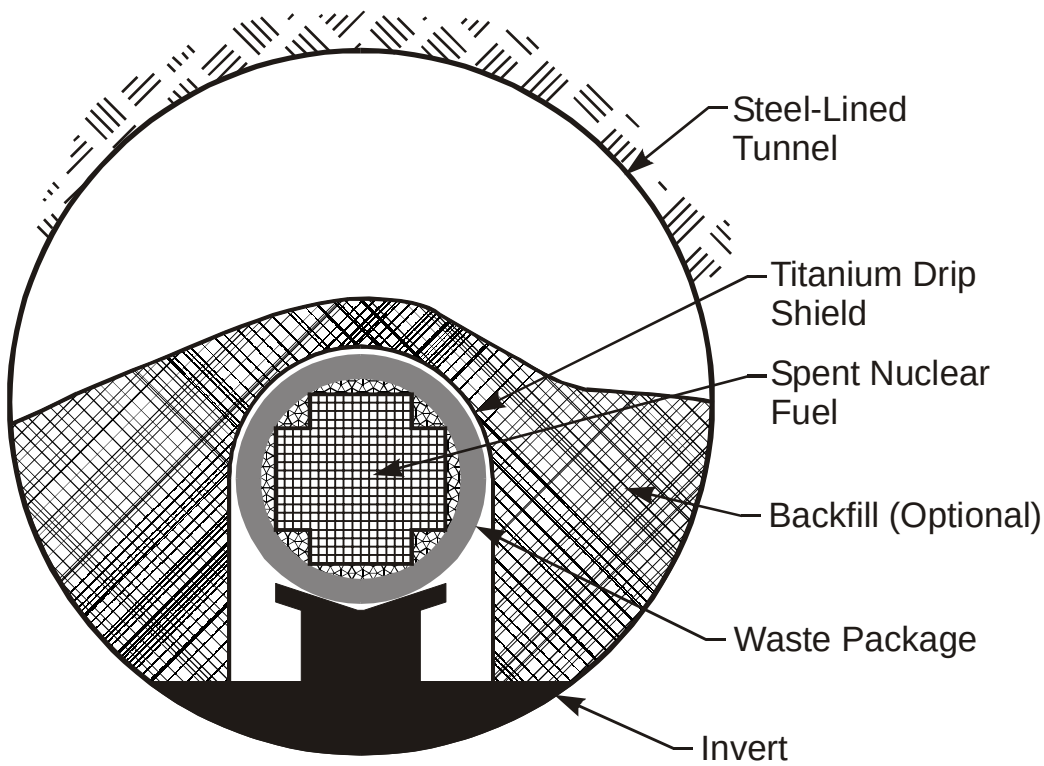
Outline

- **Concept description**
- **Benefits of using DUO_2 fill**
- **Waste package (WP) degradation behavior**
  - **Stage I: Corrosion of exterior WP**
  - **Stage II: Failure of internal structure**
  - **Stage III: Oxidation and dissolution of SNF**
  - **Stage IV: Dissolution and transport of radionuclides**
- **Conclusions**

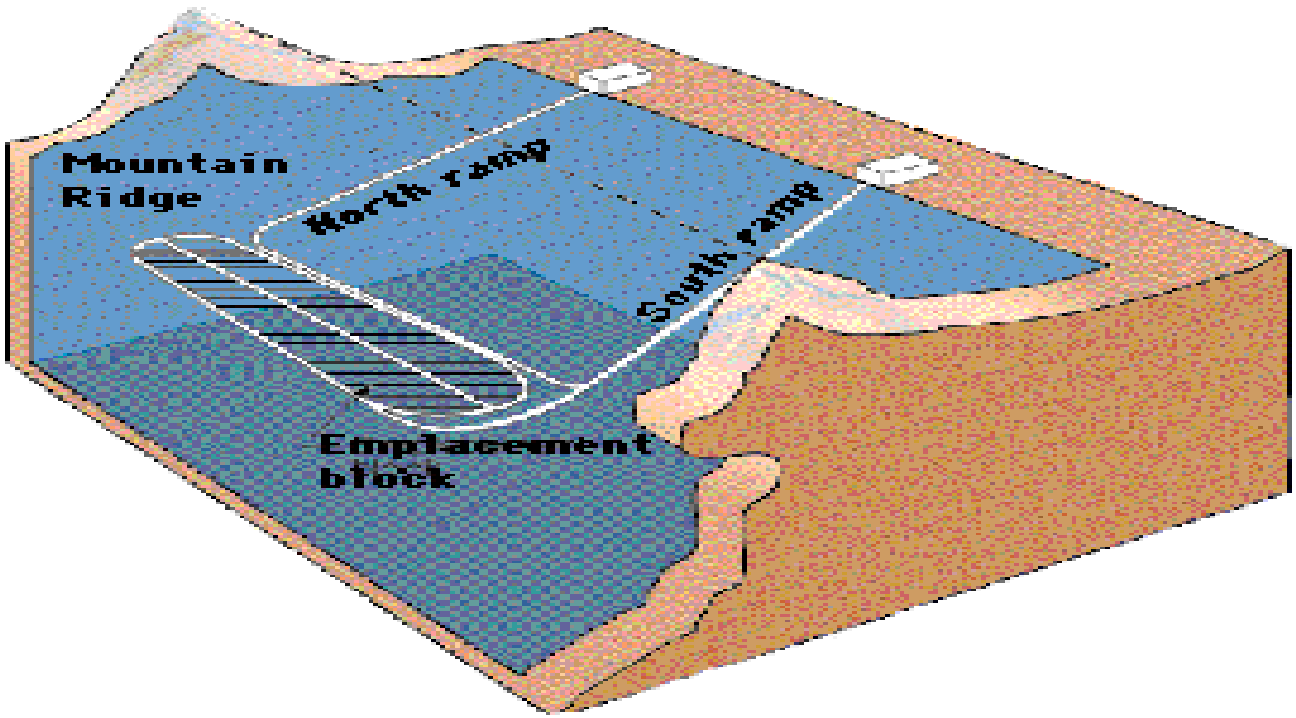
Waste Package Loading Sequence



Yucca Mountain Waste Package System



Yucca Mountain (YM) Repository



Benefits of Using DUO_2

There Are Four Potential Benefits of Using DUO_2 in a Repository WP

- **Reduce potential for long-term nuclear criticality in the repository**
- ***Reduce radionuclide release rates from SNF WPs (subject of paper)***
- **Provide WP shielding**
- **Dispose of excess DU (one-half of the inventory)**

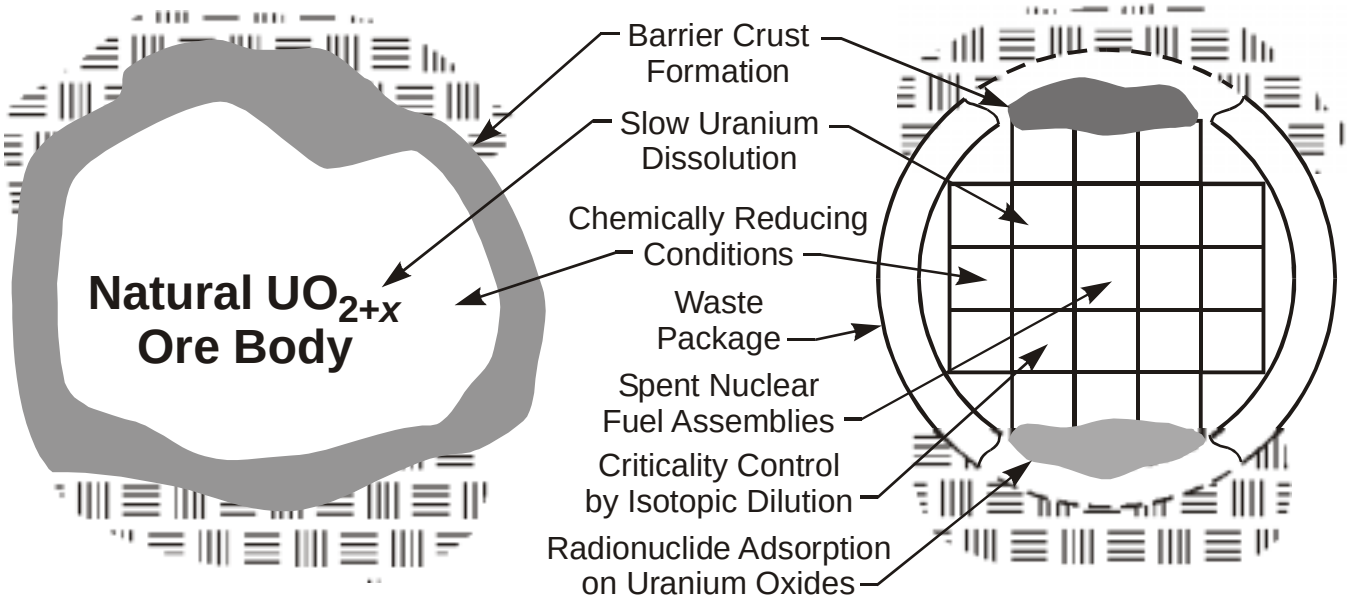
Delaying WP Radionuclide Releases Reduces Such Releases by Allowing Time for Radioactive Decay

- **Delay by preservation of SNF UO_2**
  - **Radionuclides are primarily incorporated into SNF UO_2 pellets**
  - **Radionuclides can not escape until SNF UO_2 pellets degrade**
- **Delay by retarding radionuclide movement**
  - **Minimize groundwater flow**
  - **Retard radionuclide migration in groundwater**

UO_2 Ore Deposits Have Existed for Long Periods of Times in Environments Similar to Yucca Mountain (YM)

- **UO_2 is chemically unstable in oxidizing environments such as YM**
- **SNF UO_2 oxidation will release radionuclides**
- **UO_2 ore deposits exist in such environments by sacrificial chemical reactions on the outer edges of the ore deposits**
- **The same mechanisms can be used to preserve SNF UO_2 with DUO_2 in the WP**

Some Uranium Ore Deposits Have Remained Intact for Millions of Years: The Same Mechanisms May Protect SNF with DUO_2 Fill



Impact of DUO_2 Fill on Waste Package Degradation

Waste Package Degradation Goes Through Four Stages

- **Stage I: Corrosion of exterior WP**
- **Stage II: Failure of internal structure**
- **Stage III: Oxidation and dissolution of SNF**
- **Stage IV: Dissolution and transport of radionuclides**

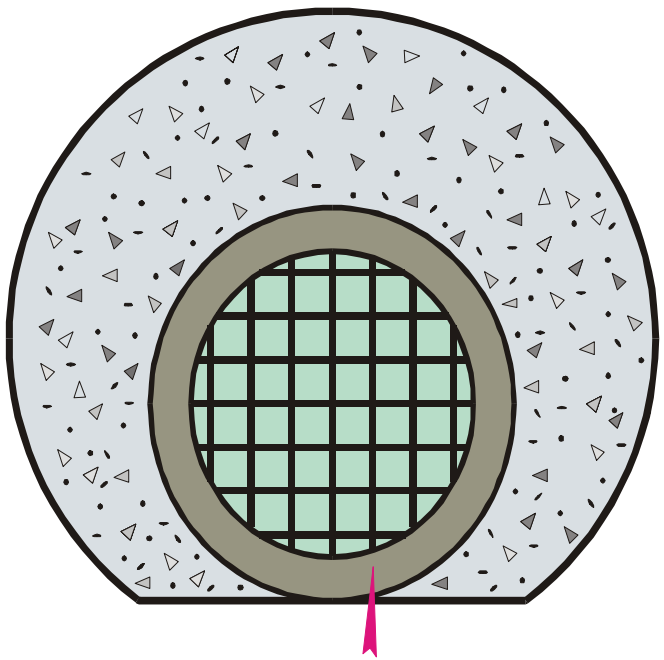
Impact of DUO_2 Fill on Waste Package Degradation

Stage I: Corrosion of exterior WP

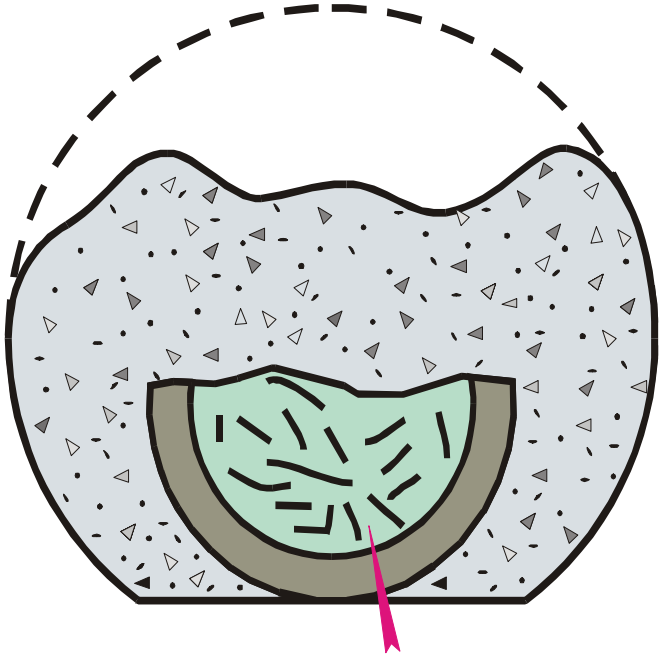
Stage I: Corrosion of Waste Package

- **No migration of SNF radionuclides occurs until after the WP fails**
- **Fill can provide internal structural support to delay WP failure (more than one-half of the WP interior is empty space)**
  - **WP walls thin with time**
  - **Tunnel collapse stresses WP**
- **Canada demonstrated the capabilities of ductile WPs supported by fill (not DUO_2)**

Fills Provide Internal Support That Delays WP Collapse with Loss of Barriers Against Groundwater Contact with SNF

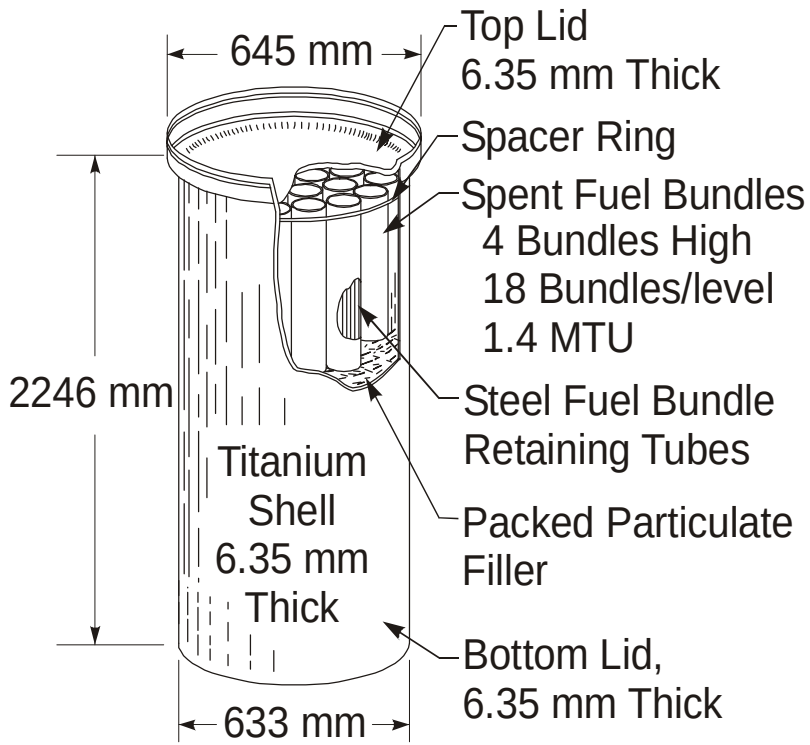


Intact Waste Package
or Waste Package with Fill
(High Resistance to Collapse)

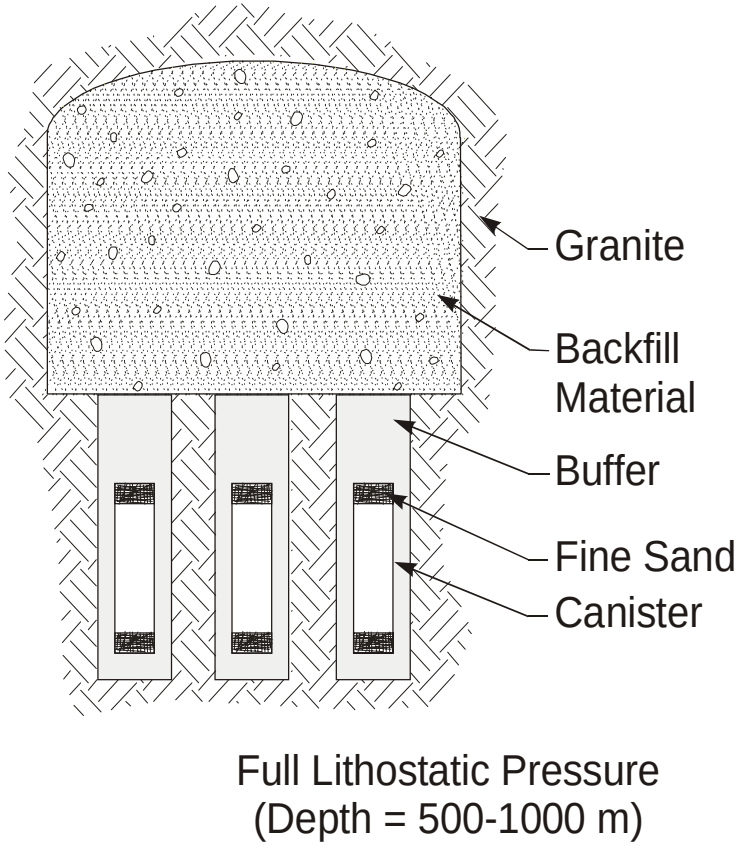


Failed Waste Package
(Collapse With No
Internal Support)

Canadian Thin-Wall (0.635 cm) WP for CANDU SNF



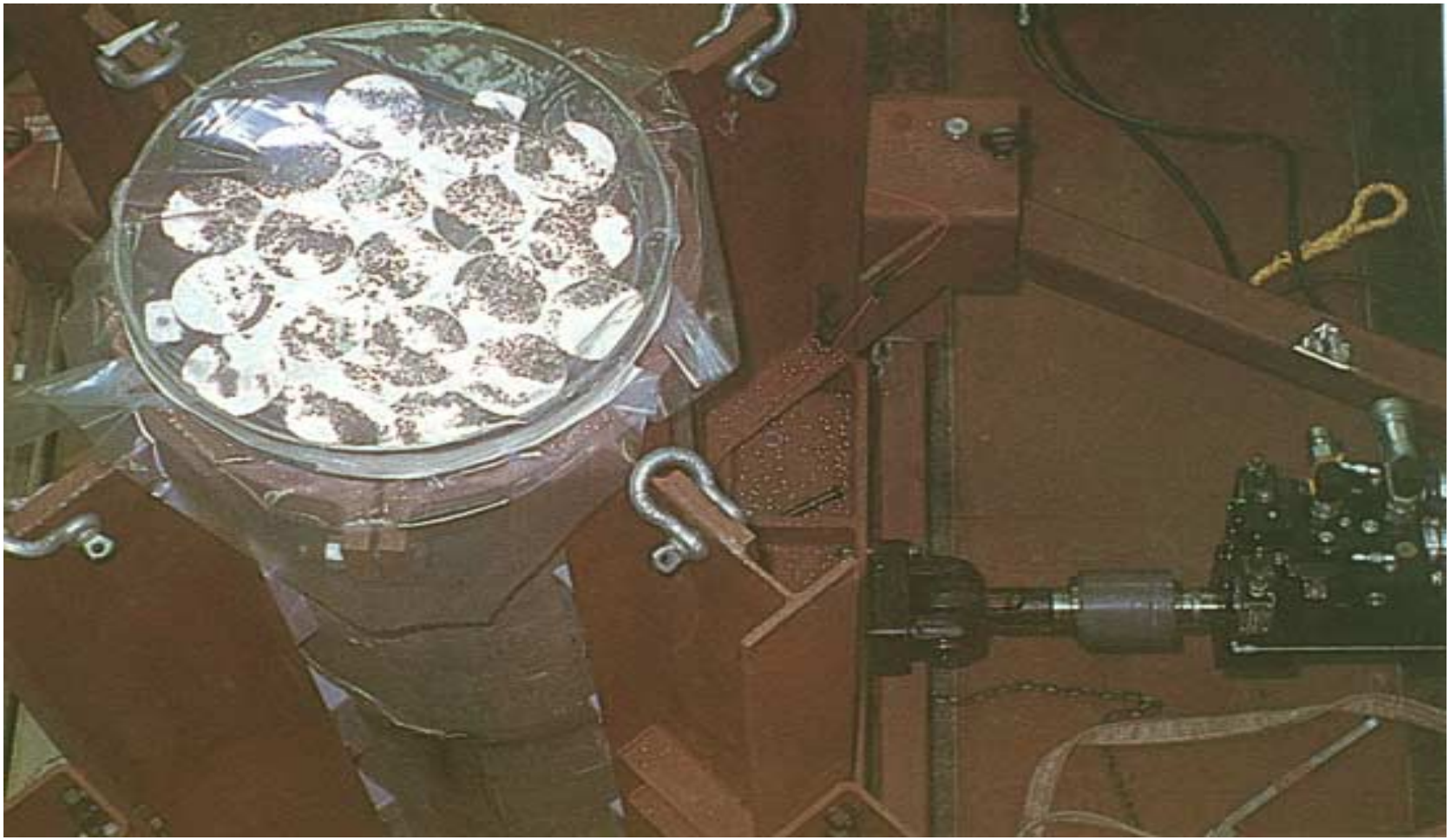
Canister Concept



Repository Disposal Concept

Canadian WP with Fill and Dummy SNF

(Courtesy of AECL)



Welding of Canadian WP (Courtesy of AECL)

Hydrostatic Test Chamber for SNF WPs
(Courtesy of AECL)

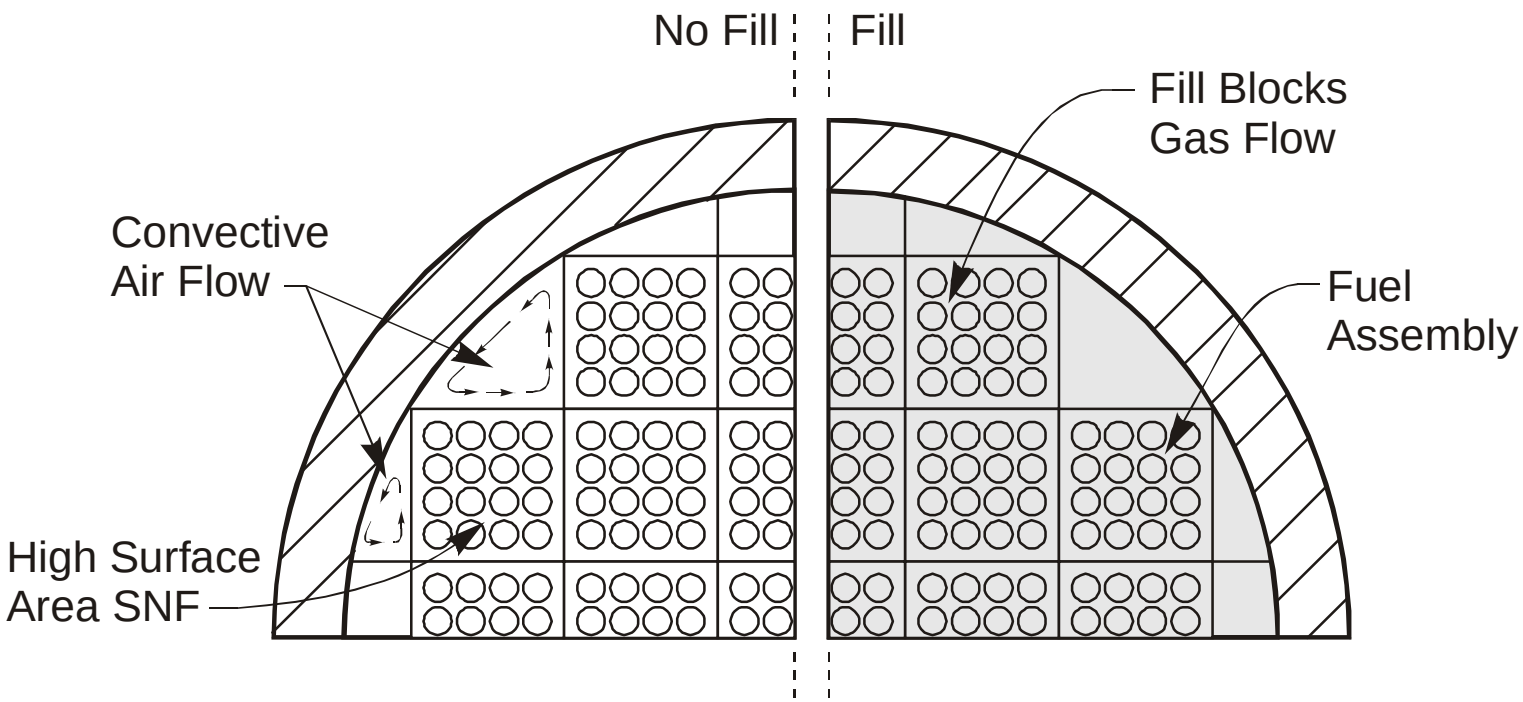
Impact of DUO_2 Fill on Waste Package Degradation

Stage II: Failure of internal structure

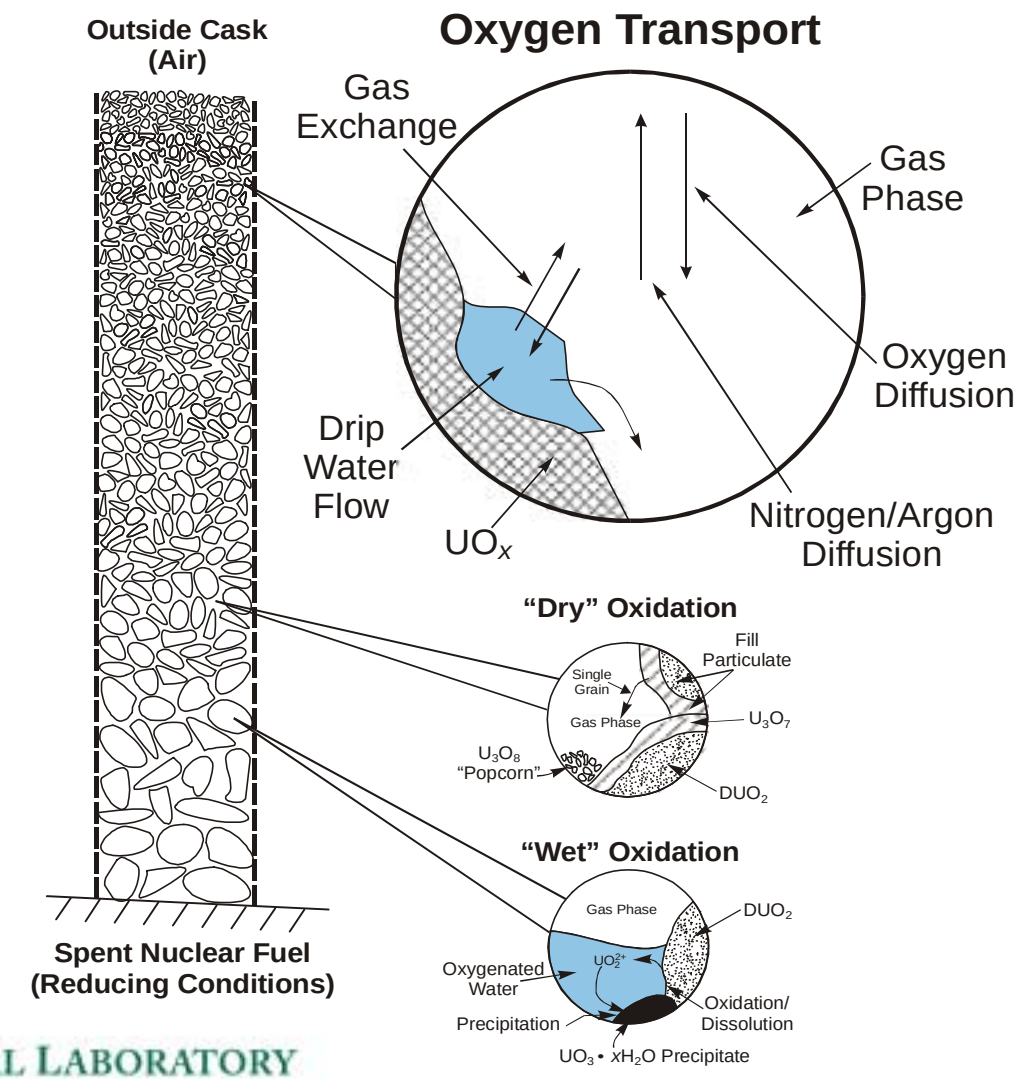
Fill Materials Can Strongly Impact Internal WP and SNF Corrosion Rates

- **No Fill**
  - **Large SNF void spaces (>50 vol %) allow rapid transport of air and water in a WP**
  - **High-surface-area SNF allows rapid corrosion**
- **Fill**
  - **Oxygen transport into WP limited by diffusion (slow process)**
  - **DUO_2 fill blocks gas flow**

Rapid Internal Degradation Occurs in WPs Without Fill Because of Rapid Air and Water Movement



Oxygen Transport to SNF in a WP With Fill Is by Gas-Phase Diffusion—A Slow Process



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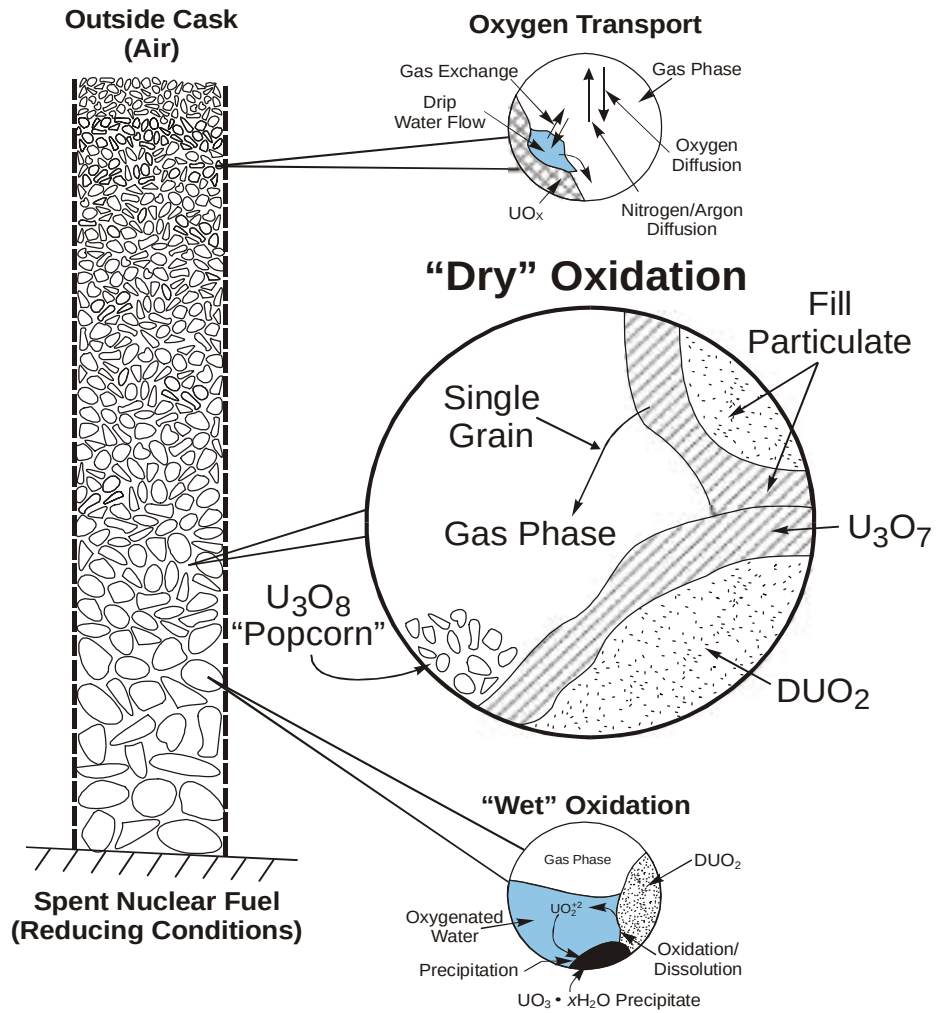
Impact of DUO_2 Fill on Waste Package Degradation

Stage III: Oxidation and dissolution of SNF

DUO_2 Slows SNF WP Radionuclide Releases by Multiple Mechanisms

- **Delay by preservation of SNF UO_2**
  - **Maintain chemically reducing conditions in the WP (avoid oxidation breakup of SNF UO_2 with releases)**
    - **DUO_2 particulates preferentially react with oxygen**
    - **DUO_2 oxidation products block WP oxygen diffusion**
  - **Saturate groundwater with DUO_2 to slow SNF UO_2 dissolution**
- **Delay by retarding radionuclide movement**
  - **Reduce groundwater flow through SNF and WP**
  - **Retard radionuclide transport in groundwater**

Dry DUO_2 Oxidation Increases the Volume of the Oxide, Breaks Fill Particulates into Smaller Grain-Size Particles, and Blocks Flow



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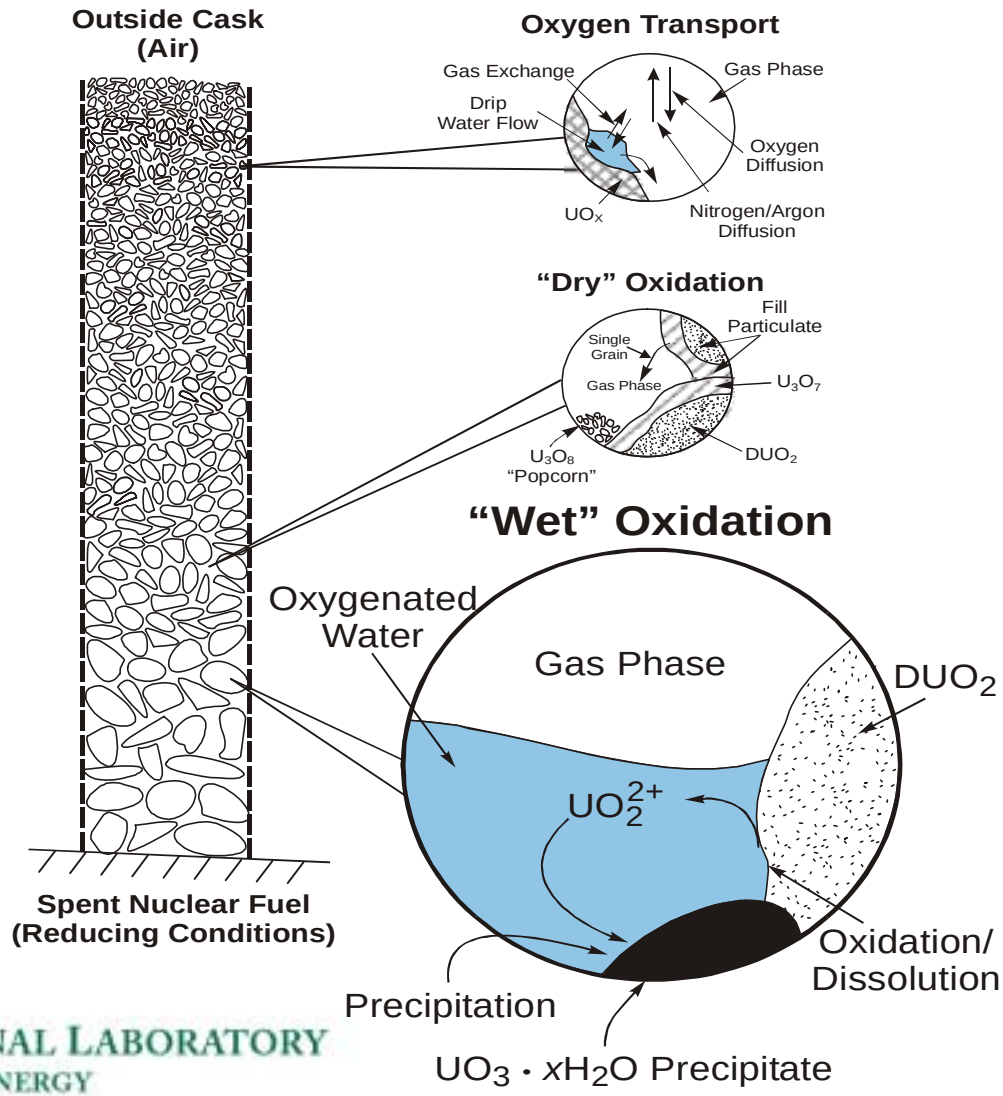
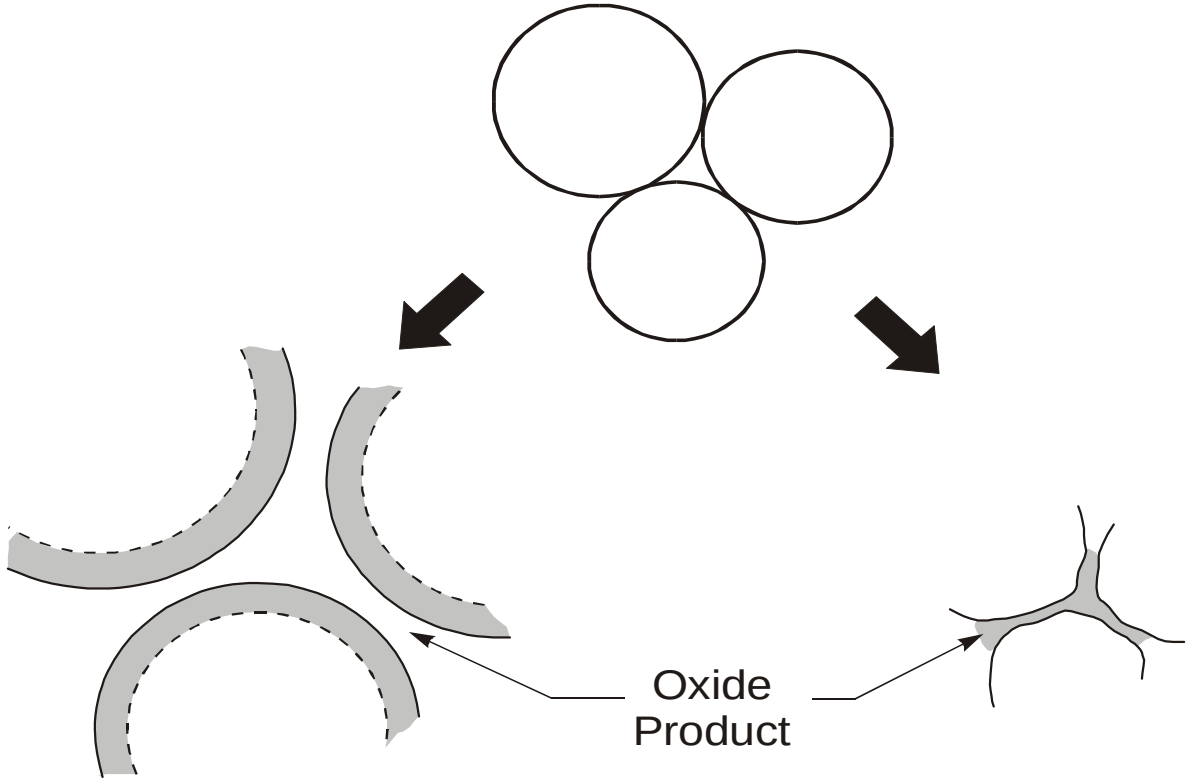
Wet Oxidation Is a Multistep Process: Oxidation, Dissolution, and Precipitation with a Volume Increase To Block Oxygen And Water Transport



DUO_2 Geochemistry Is Expected to Block Oxygen and Groundwater Flow in Fill

- **Fill expansion can increase or decrease bed permeability to gas and liquid transport**
- **DUO_2 oxidation occurs by two mechanisms**
  - **Dry oxidation**
  - **Wet oxidation**
- **Expected DUO_2 oxidation mechanisms block oxygen and groundwater transport**

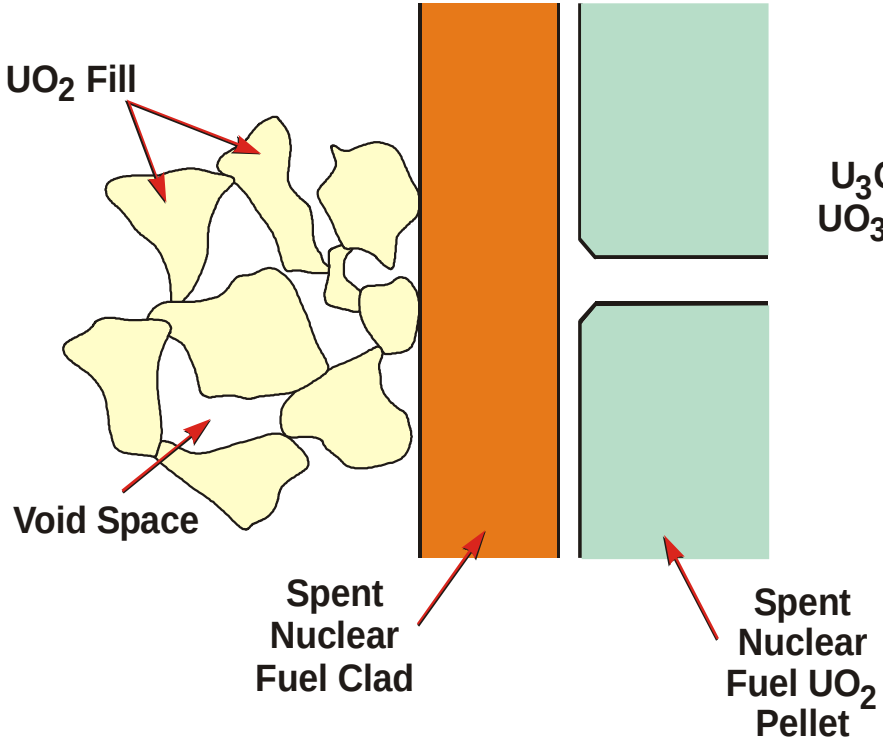
Oxidation Can Expand or Fill Void Spaces (Block Fluid Movement): DUO_2 Is Expected to Fill Spaces



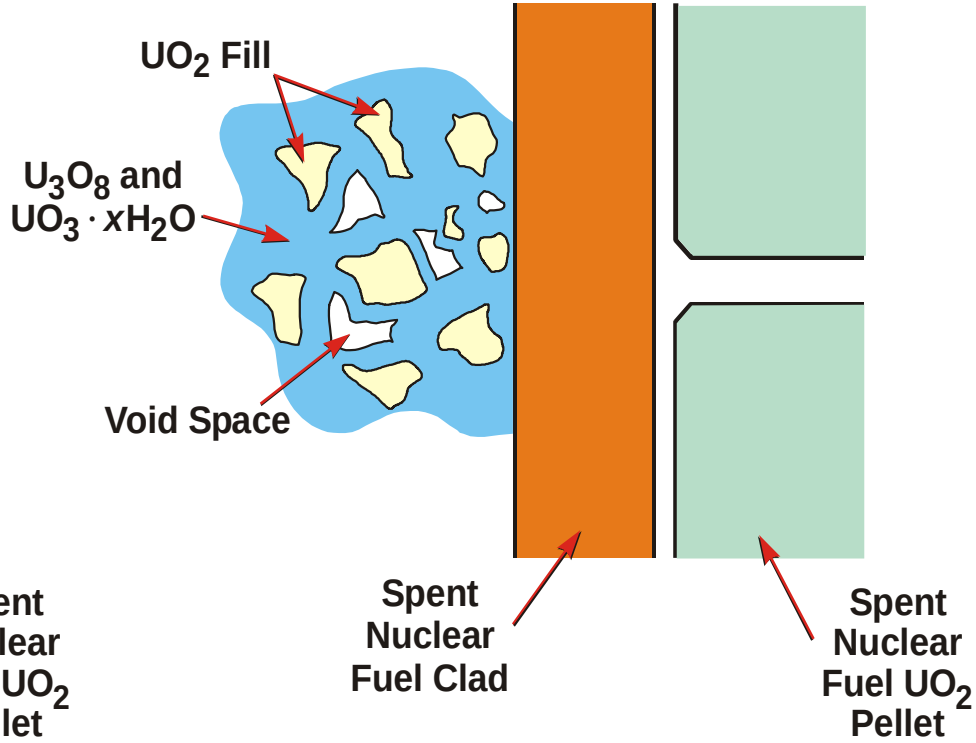
Expanding Voids **Blocked Voids**

DUO_2 Is Expected To Reduce Fluid (Oxygen and Groundwater) Flow

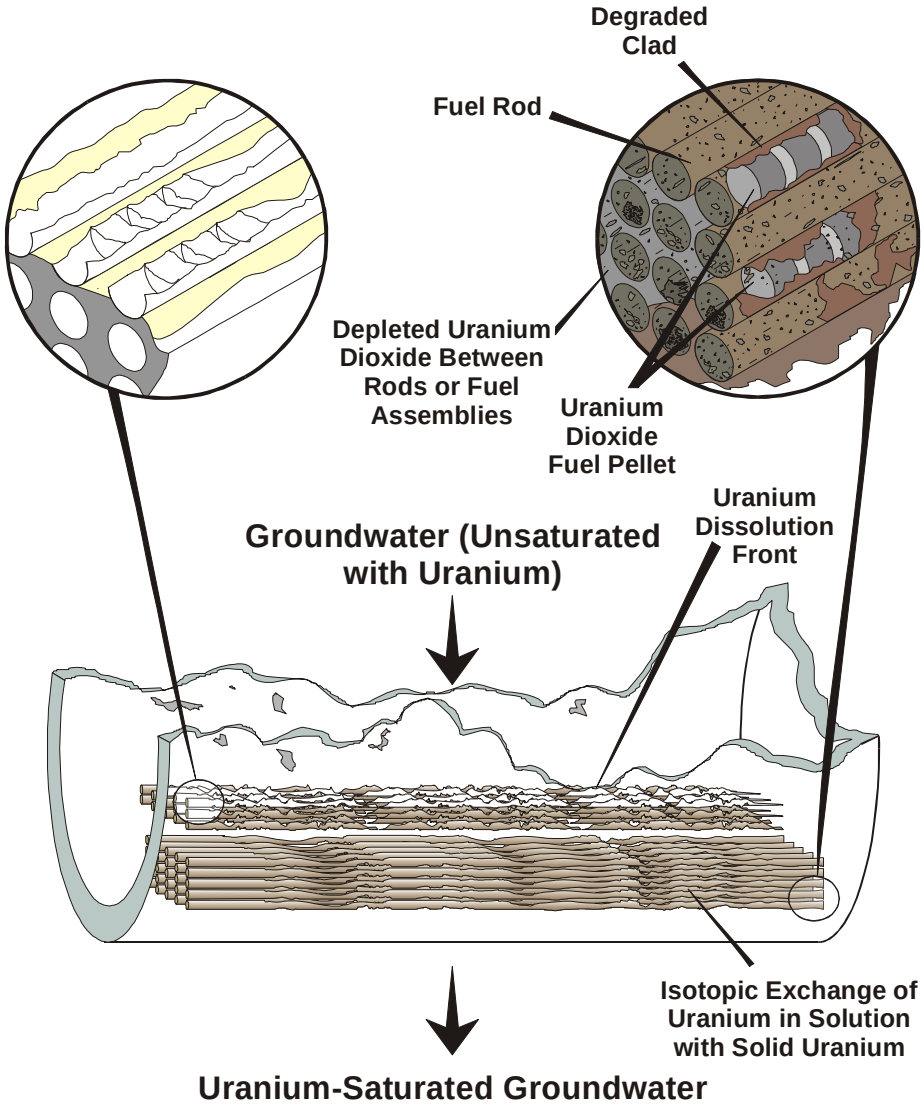
Initial Conditions



Conversion of UO_2 to Higher Oxides



If Groundwater Flow Is Shut Down, Congruent Dissolution Occurs with Very Slow Release of Radionuclides



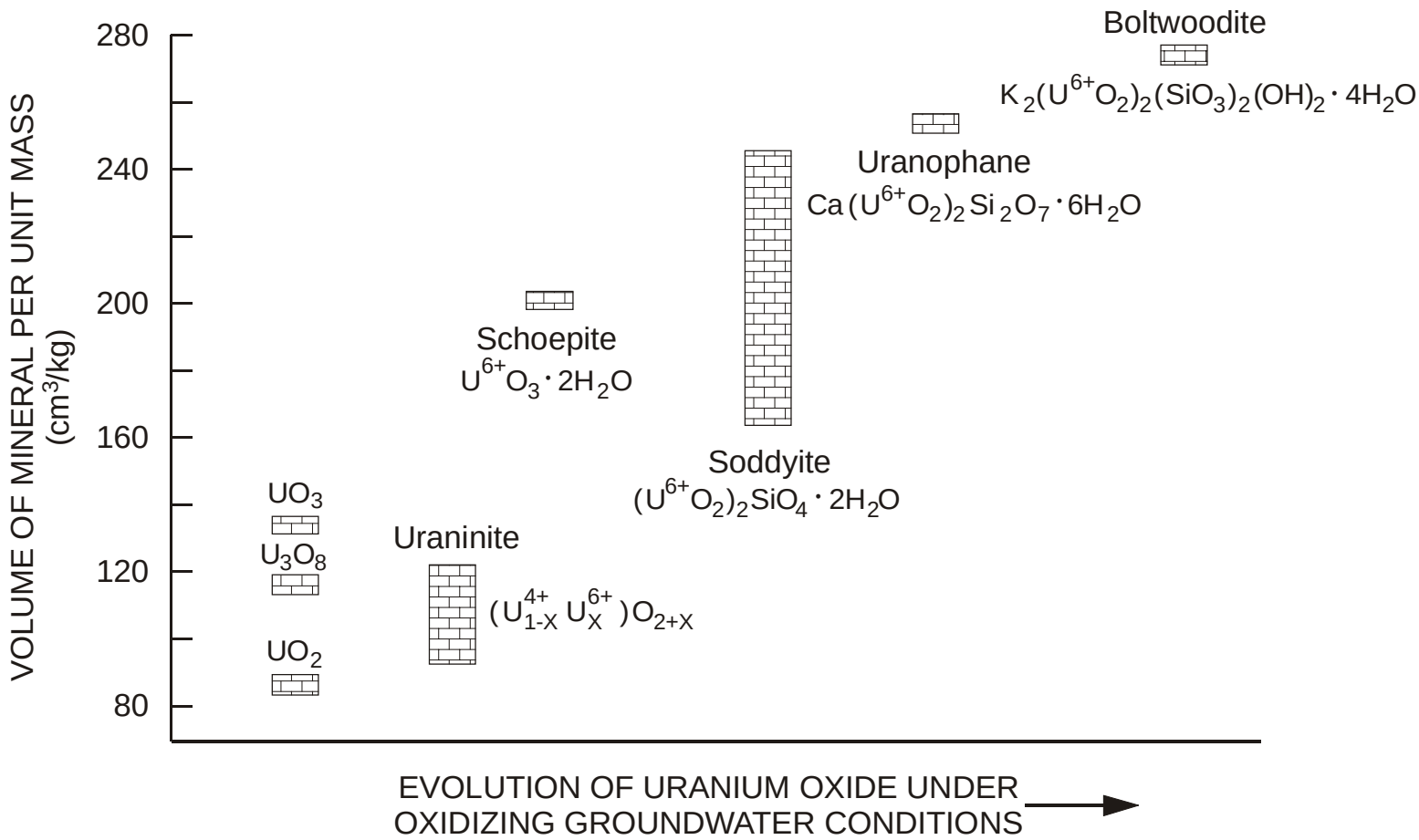
Impact of DUO_2 Fill on Waste Package Degradation

Stage IV: Dissolution and transport of radionuclides

Hydrated DU Oxidation Products Will Delay WP Radionuclide Releases

- **Heavy metal hydrated oxides sorb and ion-exchange with many radionuclides**
- **Limited data on hydrated uranium oxides show this effect**
- **Data is not currently available for the radionuclides of greatest interest under the expected groundwater conditions**

Evolution of Uranium Oxides over Time to Hydrated Uranium Silicates Tends To Block Air and Groundwater Flow



Conclusions

- **The addition of a DUO_2 fill significantly alters the degradation of an SNF WP**
- **The potential exists for significant improvement in WP performance**
  - **Natural analogues (uranium deposits) suggest fills will improve WP performance**
  - **Mechanistic understanding of uranium oxidation supports this concept**
- **Many uncertainties require analysis and experiments**