Wpf:ans99du5/a

ADVANCED SPENT FUEL WASTE PACKAGE FILL MATERIAL: DEPLETED URANIUM DIOXIDE

Charles W. Forsberg

Chemical Technology Division
Oak Ridge National Laboratory*
Oak Ridge, Tennessee 37831-6180
Tel: (423) 574-6783
Fax: (423) 574-9512
Email: forsbergcw@ornl.gov

Session: Alternative Analysis for Geological Disposal
American Nuclear Society 1999 Winter Meeting
Long Beach, California
November 14–18, 1999

"The submitted manuscript has been authored by a contractor of the U.S. Government under contract No. DE-AC05-96OR22464. Accordingly, the U.S. Government retains a nonexclusive, royalty free licence to publish or reproduce the published form of this contribution, or allow other to do so, for U.S. Government purposes."

---

*Managed by Lockheed Martin Energy Research Corp., under contract DE-AC05-96OR22464 for the U.S. Department of Energy.

Outline

- Repository Goals
- Potential impacts of depleted uranium dioxide (DUO_2) fill in Spent Nuclear Fuel (SNF) waste packages on repository performance
- Engineering approach

The Objective Of A Repository Is To Isolate Hazardous Radionuclides Until They Decay To Low Levels

- The hazardous radionuclides are primarily incorporated into the UO_2 fuel pellets in SNF assemblies
- If the UO_2 can be preserved, radionuclide releases from the WP are low
- High repository performance can be assured if SNF UO_2 integrity is assured

Potential Impacts of DUO_2 Fill In SNF Waste Packages On Repository Performance

Uranium Dioxide In Natural Uranium Ore Deposits In Environments Similar To Yucca Mountain, On A Small Scale, Has Existed For Millions Of Years

---

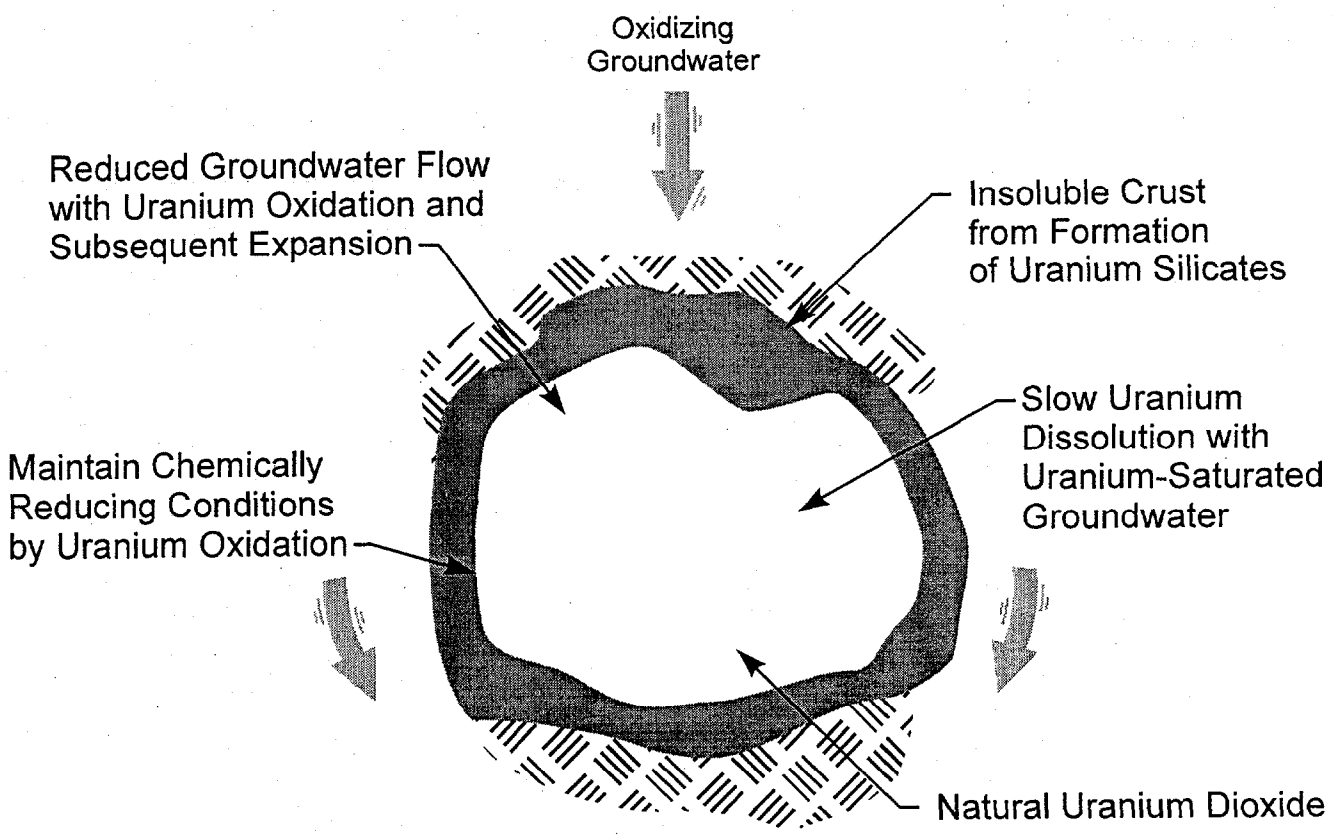
- Uranium dioxide is geologically unstable in oxidizing environments such as Yucca Mountain. Uranium dioxide oxidizes to higher oxides with the ensuing release of components trapped in the UO_2 crystal structure

- The UO_2 is protected by sacrificial chemical reactions of uranium and other chemical species on the outer edges of such deposits

- The same mechanisms can be used to preserve SNF UO_2 and reduce radionuclide releases from a failed waste package

---

ORNL DWG 99C-362

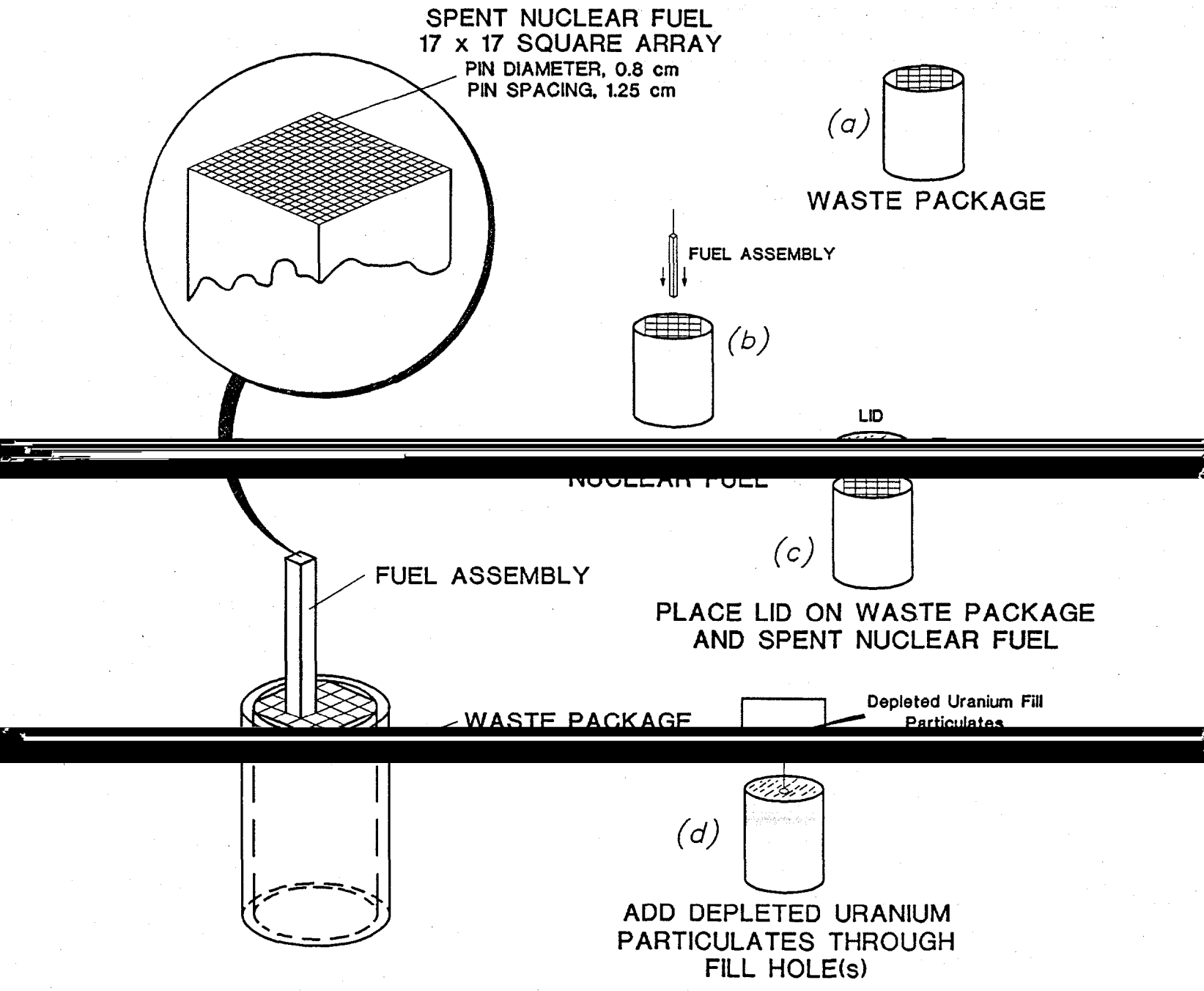


The Interiors of Natural Uranium Ore Deposits of Uranium Dioxide are Preserved for Extended Times by Degradation of Uranium Oxides on the Outside Surfaces of the Ore Deposit

An Analog Approach May Be Used To Improve SNF Repository Performance

- Fill waste packages with SNF
- Add DUO_2 particulate fill (0.5–1.0 mm) to voids inside the waste package
  - —About 3 tons of DUO_2 can be added per ton of SNF
  - —Small particle size allows filling SNF coolant channels
- Waste package becomes a man-made uranium ore deposit with the DUO_2 particulate preferentially reacting with the environment and protecting the SNF UO_2. DUO_2 preferentially reacts with groundwater:
  - —DUO_2 is a high-surface-area particulate compared to SNF UO_2
  - —SNF UO_2 is partly protected by clad

ORNL DWG 97A-78R

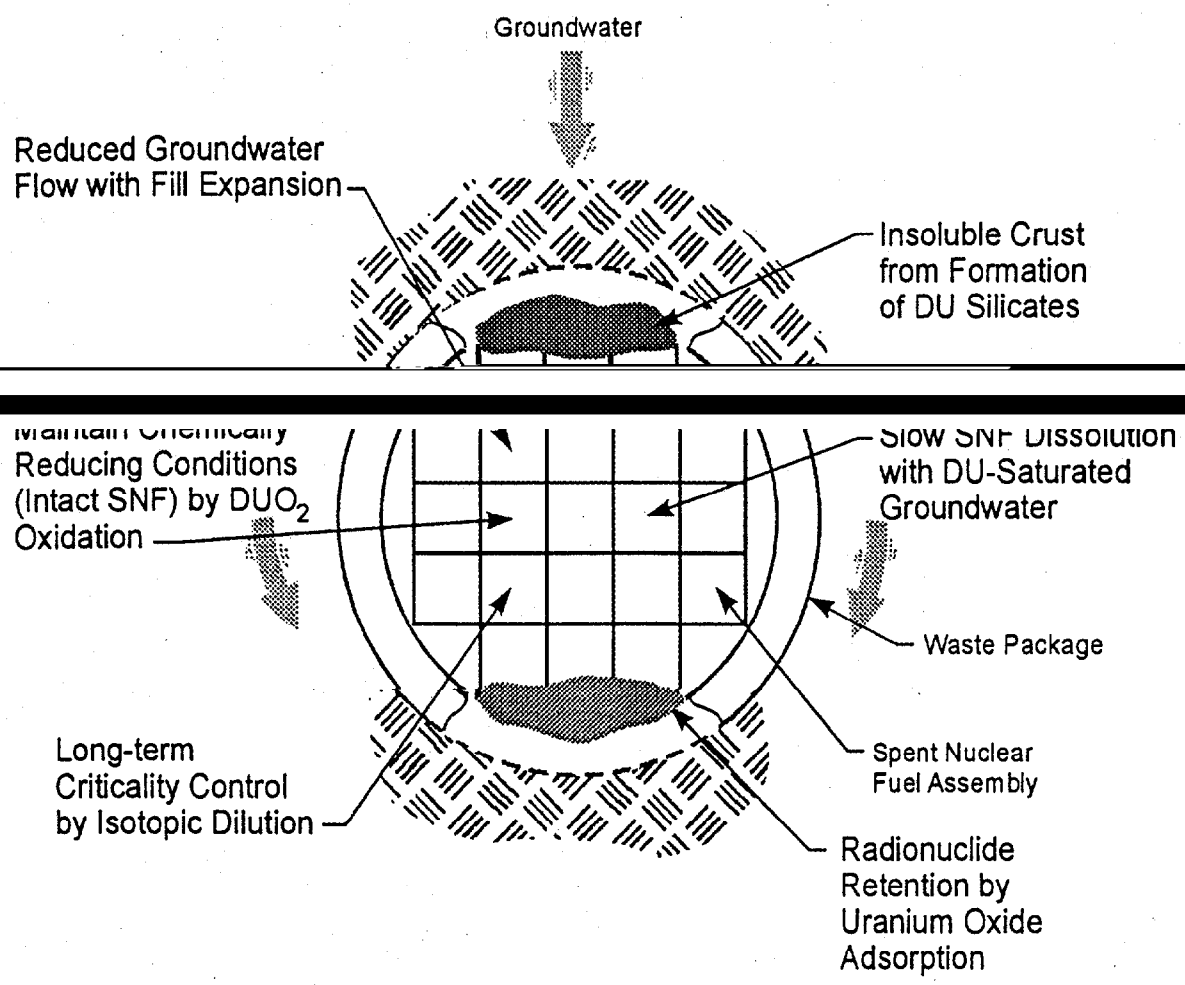


Waste Package System and
Example of Loading Sequence
for Depleted Uranium Particulates

The DUO_2 Fill Slows Radionuclide Releases After Waste Package Failure By Multiple Mechanisms

- Maintains chemically reducing conditions in the waste package
- Reduces air and groundwater movement through the waste package
- Retards radionuclide releases by ion exchange
- Saturates groundwater with uranium to slow SNF UO_2 dissolution
- Minimizes potential for nuclear criticality

ORNL DWG 99C-35



Spent Nuclear Fuel Waste Package with Depleted Uranium Dioxide Particulate Fill to Improve Repository Performance

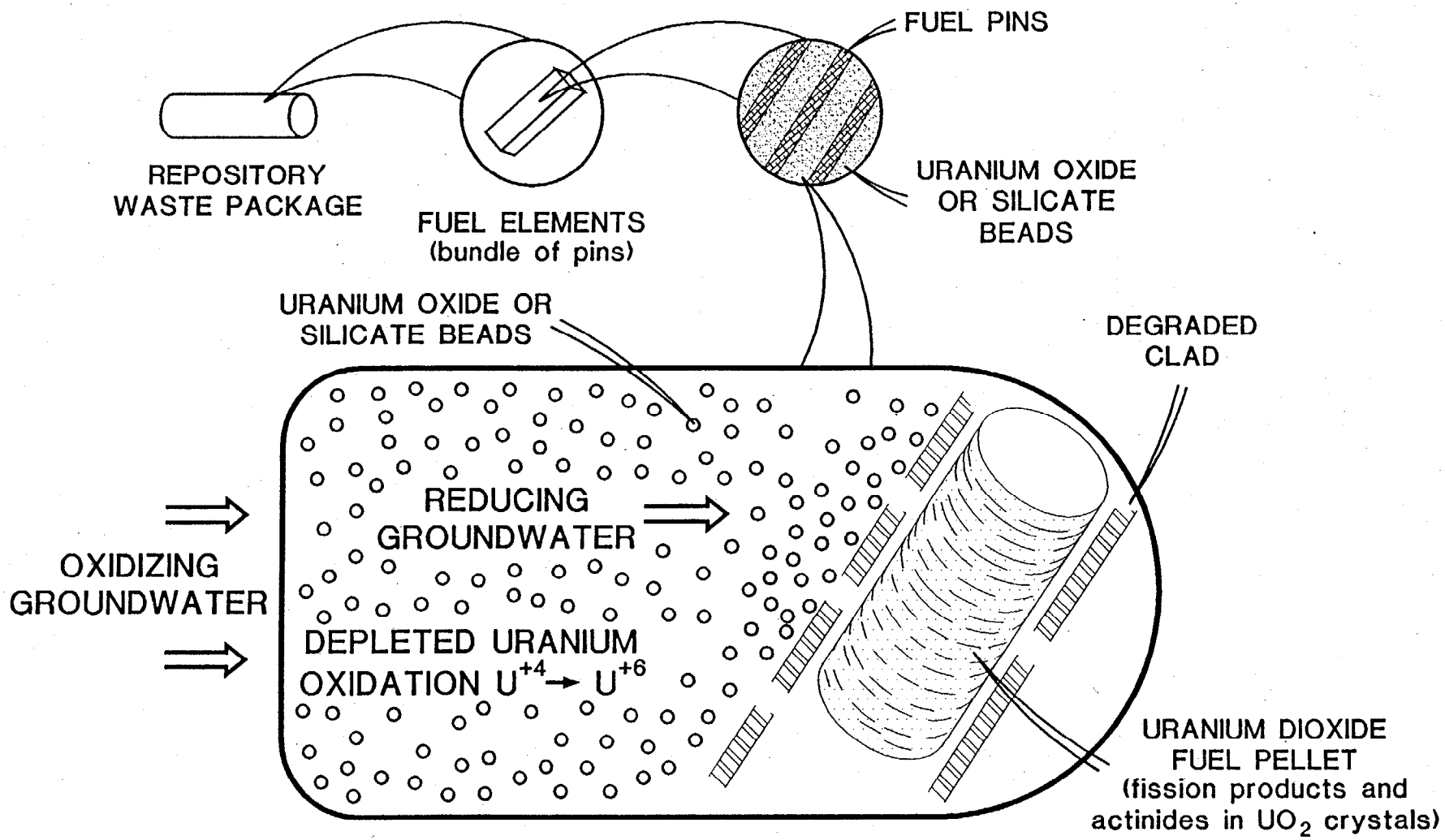
The DUO_2 Reduces Radionuclide Releases By Maintaining Chemically Reducing Conditions In The Waste Package That Prevent SNF UO_2 Degradation

---

- SNF and SNF UO_2 are chemically stable under reducing conditions
- Under oxidizing conditions, such as those at Yucca Mountain, SNF UO_2 is oxidized to U_3O_8 and $UO_3 \bullet xH_2O$ with the ensuing release of radionuclides to the groundwater
- The DUO_2 preferentially reacts with oxygen, is oxidized to U_3O_8 and $UO_3 \bullet xH_2O$, and consequently maintains chemically reducing conditions in the waste package

---

ORNL DWG 96A-333R



Depleted Uranium Oxides and Silicates in the 4+ Valence State Help Maintain Chemically Reducing Groundwater (with Low UNF Uranium Solubility) in the Waste Package

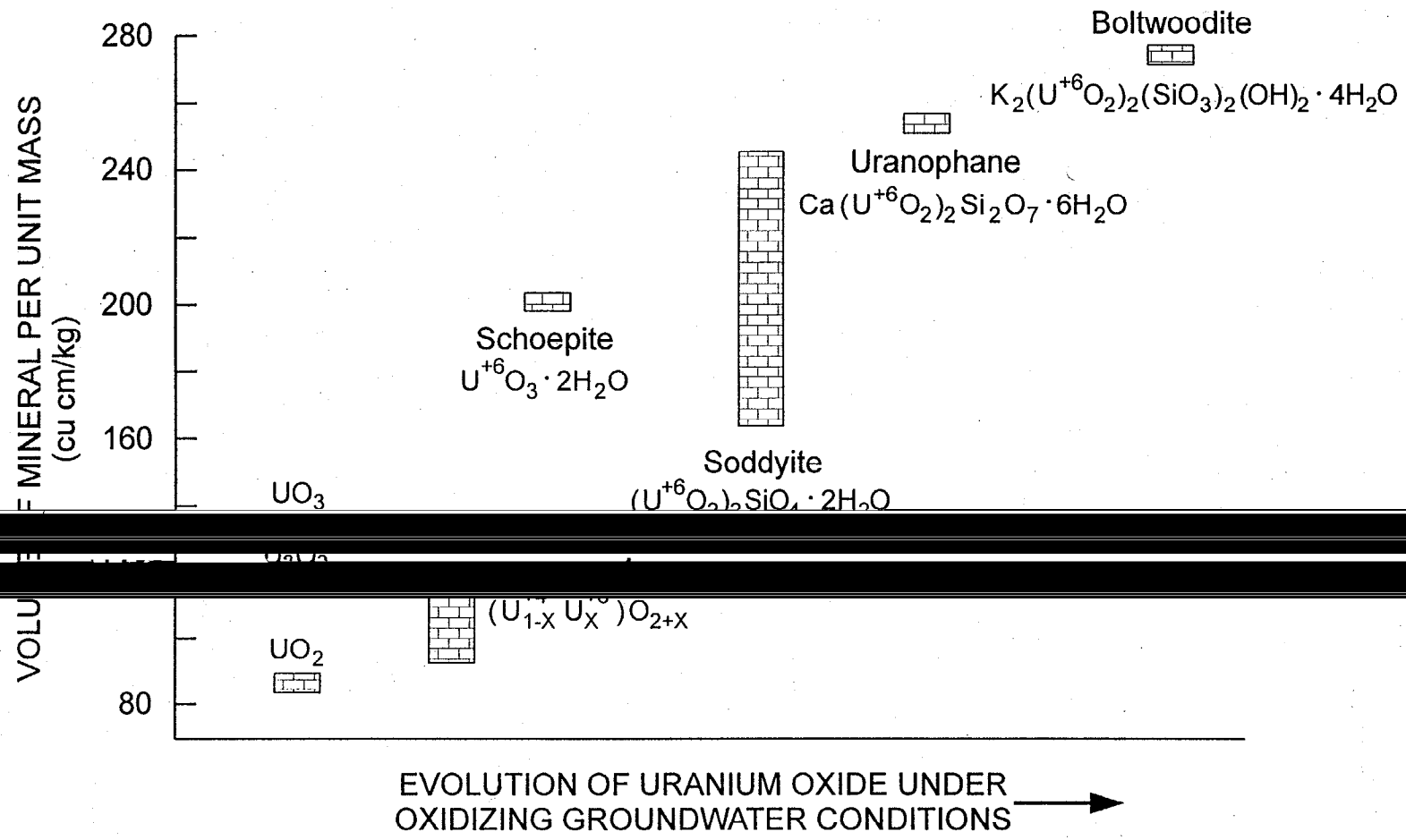
The DUO_2 Reduces Radionuclide Releases By Reducing Air And Groundwater Movement In The SNF Waste Package

- The oxidizing groundwater oxidizes the DUO_2 to U_3O_8 and $UO_3 \cdot xH_2O$
- The volume of the oxidation products are >30% than the volume of the DUO_2
- This swelling fills the interparticle void space and reduces the permeability of the waste package to air and groundwater flow

  —Oxygen transport into the SNF is slowed

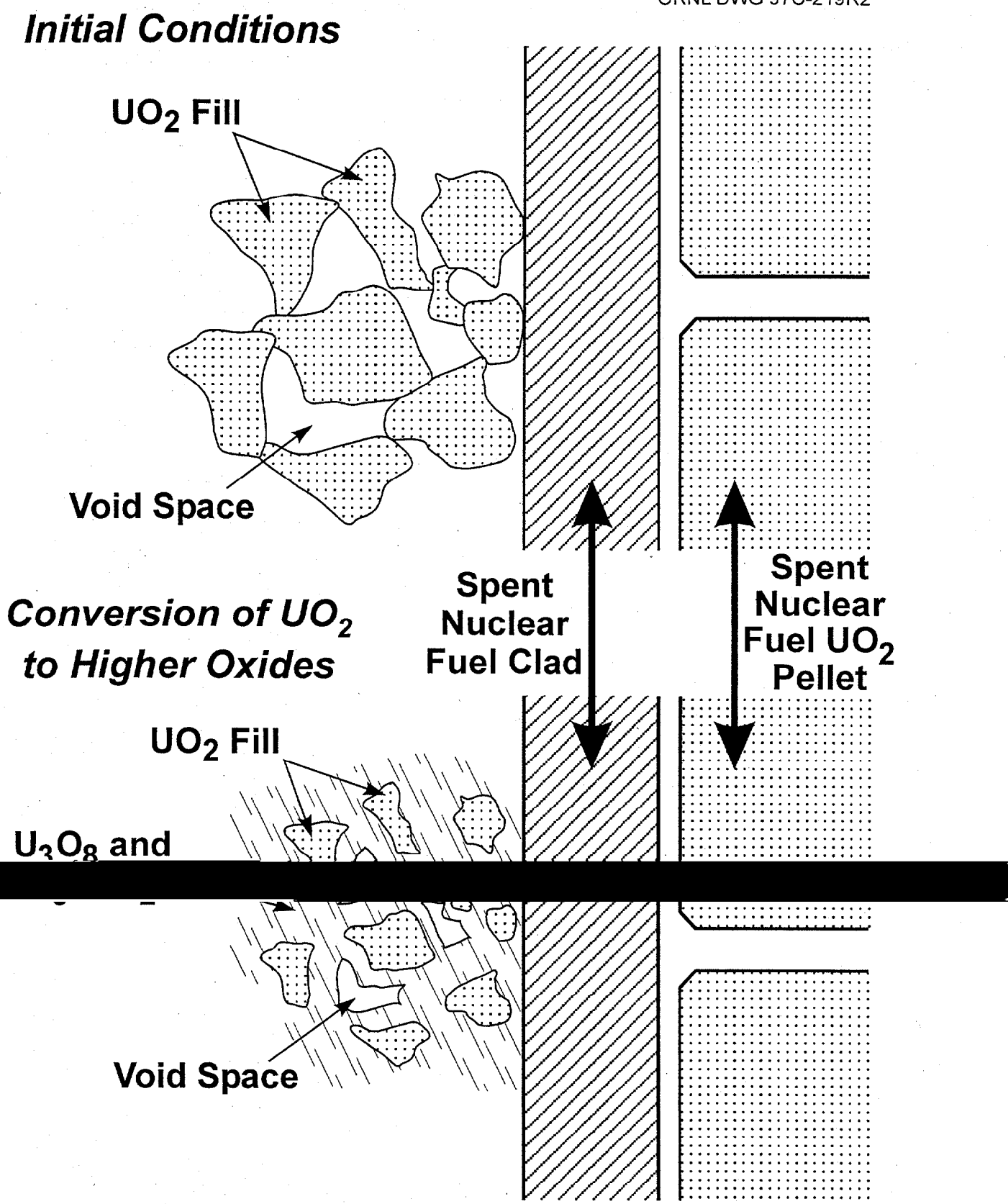
  —Groundwater flows around the waste package

ORNL DWG 96C-326R



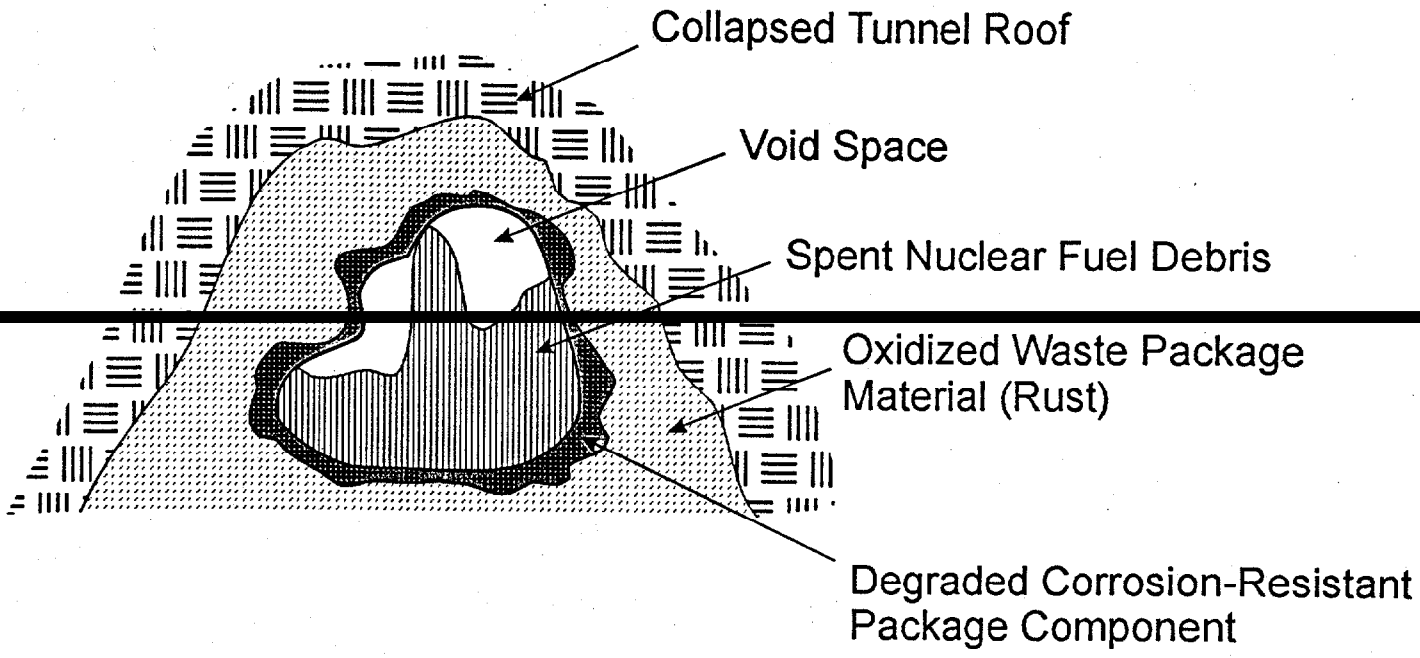
Volume of the Uranium Mineral Versus Evolution of Uranium

ORNL DWG 97C-219R2

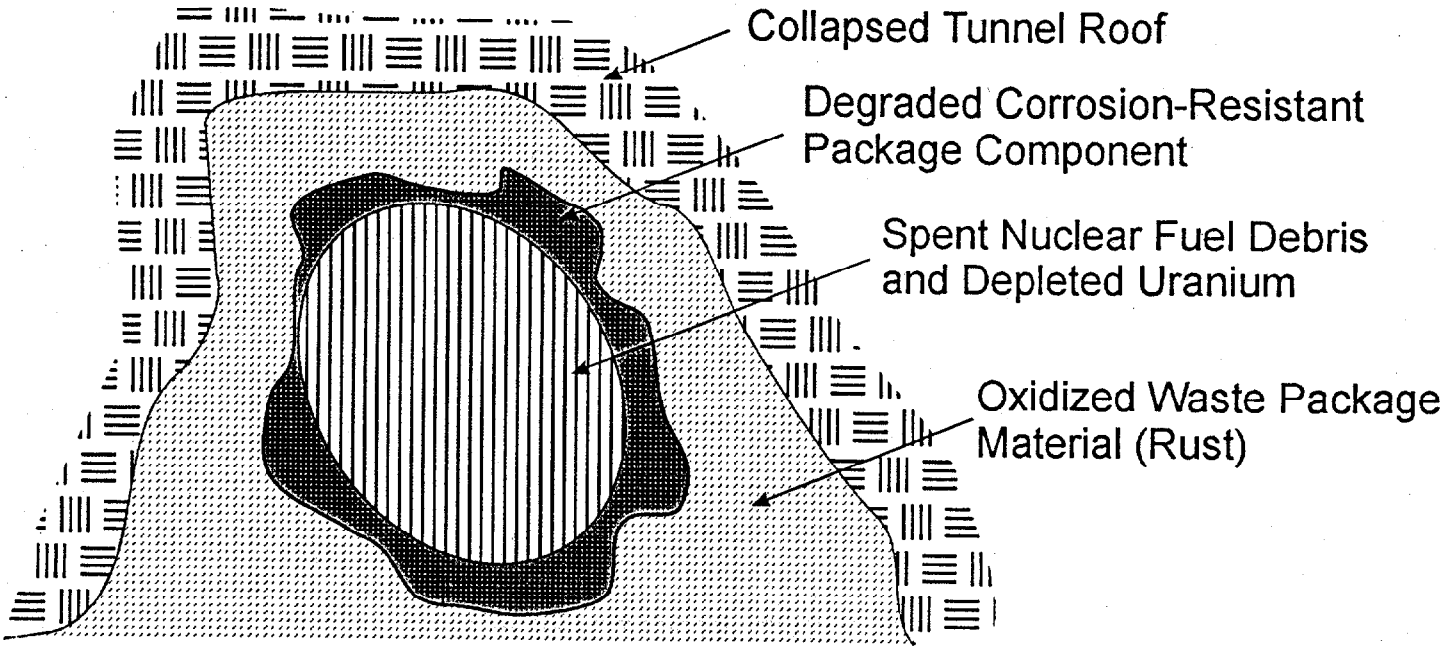


ORNL DWG 97C-221

Waste Package with No Fill (Permeable to Water Flow)



Waste Package with Fill (Low Permeability to Water Flow)



Degraded Waste Packages With and Without Internal Fill

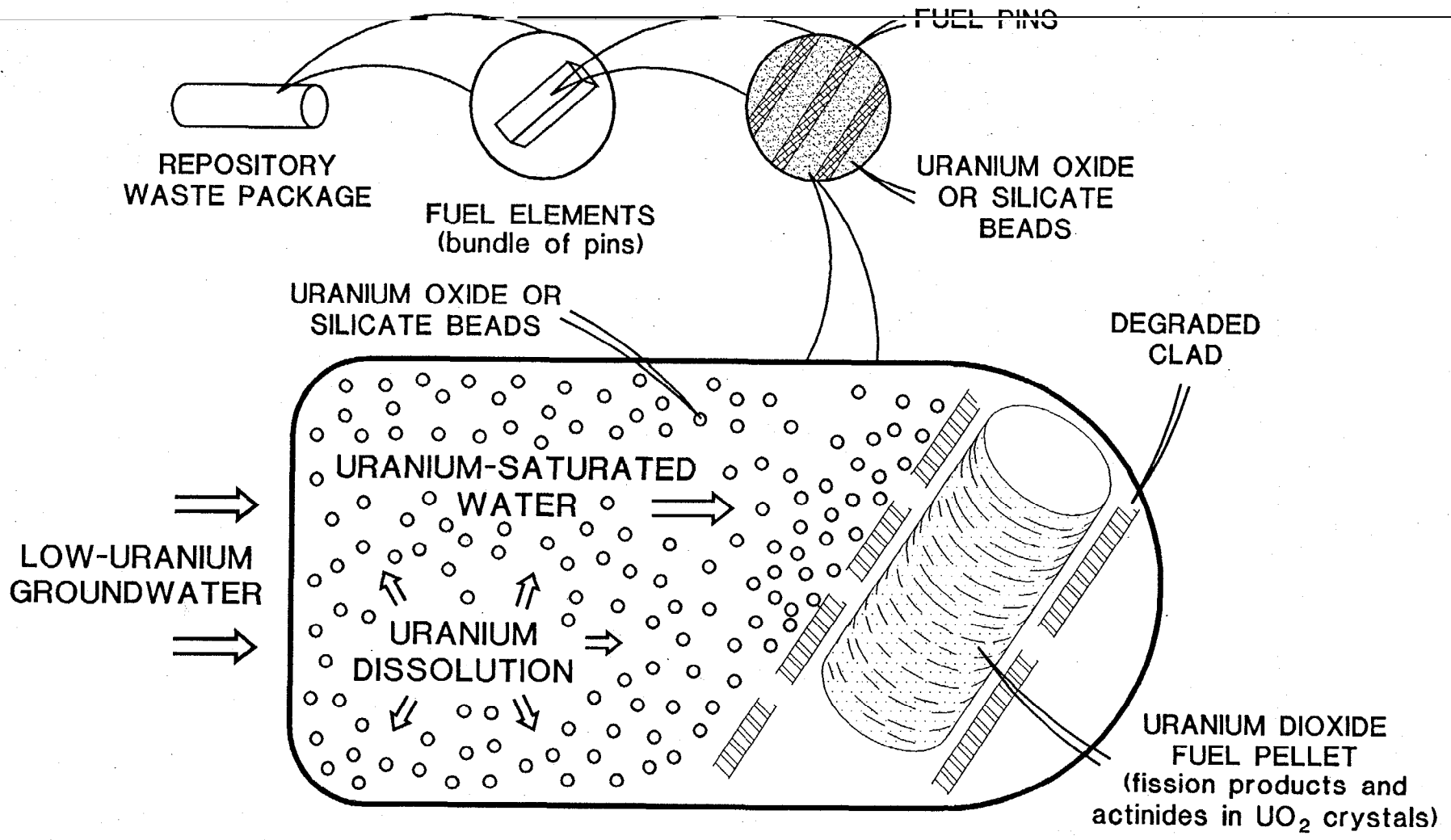
The DUO_2 Reduces Radionuclide Releases By Ion Exchange In The Waste Package

- Heavy-metal hydrated oxides act as inorganic ion-exchange materials
- Available data indicate that uranium oxides operate in a similar manner
- Migration of radionuclides such as neptunium from the SNF is expected to be slowed

The DUO_2 Reduces Radionuclide Releases By Saturating The Groundwater In The Waste Package And Slowing The SNF Uranium Dissolution Process

- The DUO_2 will saturate any water entering the waste package with uranium
- Uranium-saturated groundwater can not dissolve SNF uranium. Releases from the SNF are slowed

ORNL DWG 96A-332



Depleted Uranium Fill Saturates Waste Package Groundwater with Uranium and Thus Minimizes SNF Uranium Dioxide Dissolution

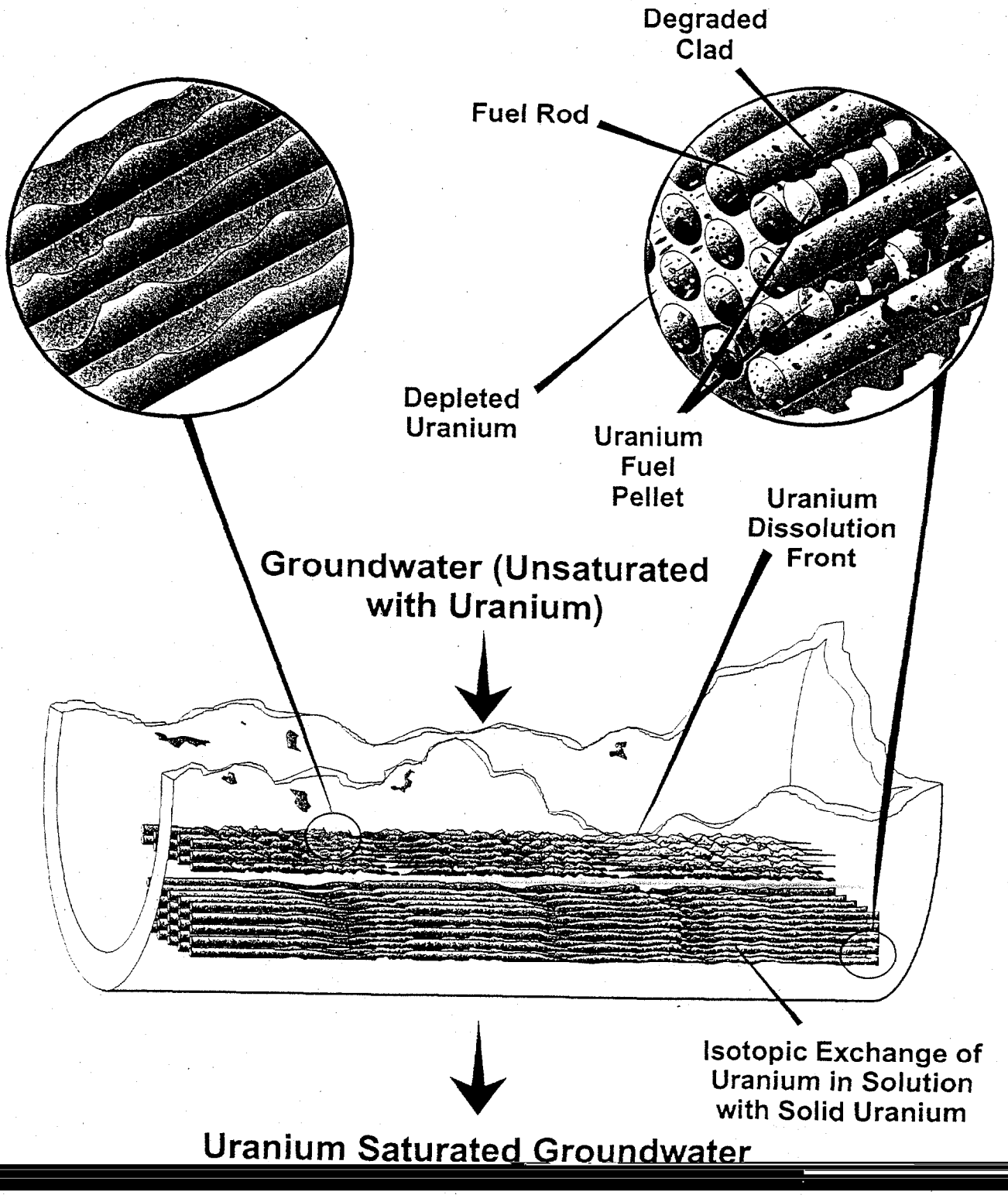
The DUO_2 Reduces Radionuclide Releases By Minimizing The Potential For Nuclear Criticality

- In the distant past, nuclear criticality has occurred in natural uranium ore bodies at fissile assays as low as 1.3 wt % ^{235}U equivalent. Nuclear criticality generates heat that can accelerate water movement and the transport of radionuclides

- The average fissile assay of LWR SNF is ~ 1.5 wt % ^{235}U equivalent; if the uranium dissolves, migrates, and precipitates in the proper geometry, nuclear criticality is possible

- The DUO_2 fill lowers the average waste package fissile content to <0.6 wt % ^{235}U equivalent—nuclear criticality can not occur at these fissile contents

ORNL DWG 98C-65



Isotopic Dilution of Spent Nuclear Fuel Uranium with Depleted Uranium

Engineering Approach

The Basic Technology For UO_2 Particulate Fill Addition To Waste Packages Exists

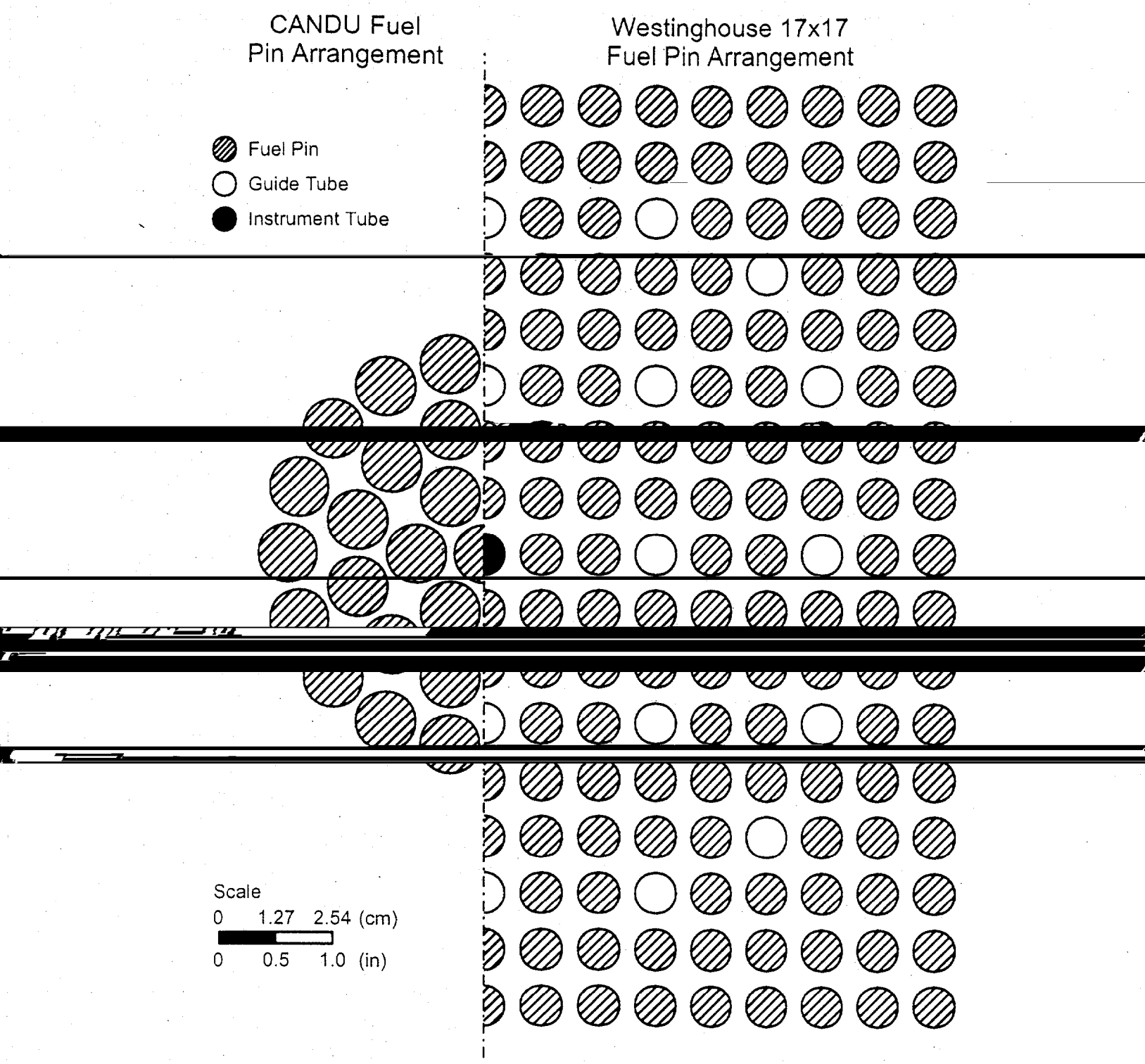
- Several countries (Canada, Spain, etc.) plan to use fill materials in waste packages
- The Canadians have conducted multiple, full-scale fill tests.

  — The technology has been successfully tested on multiple fills but not on DUO_2

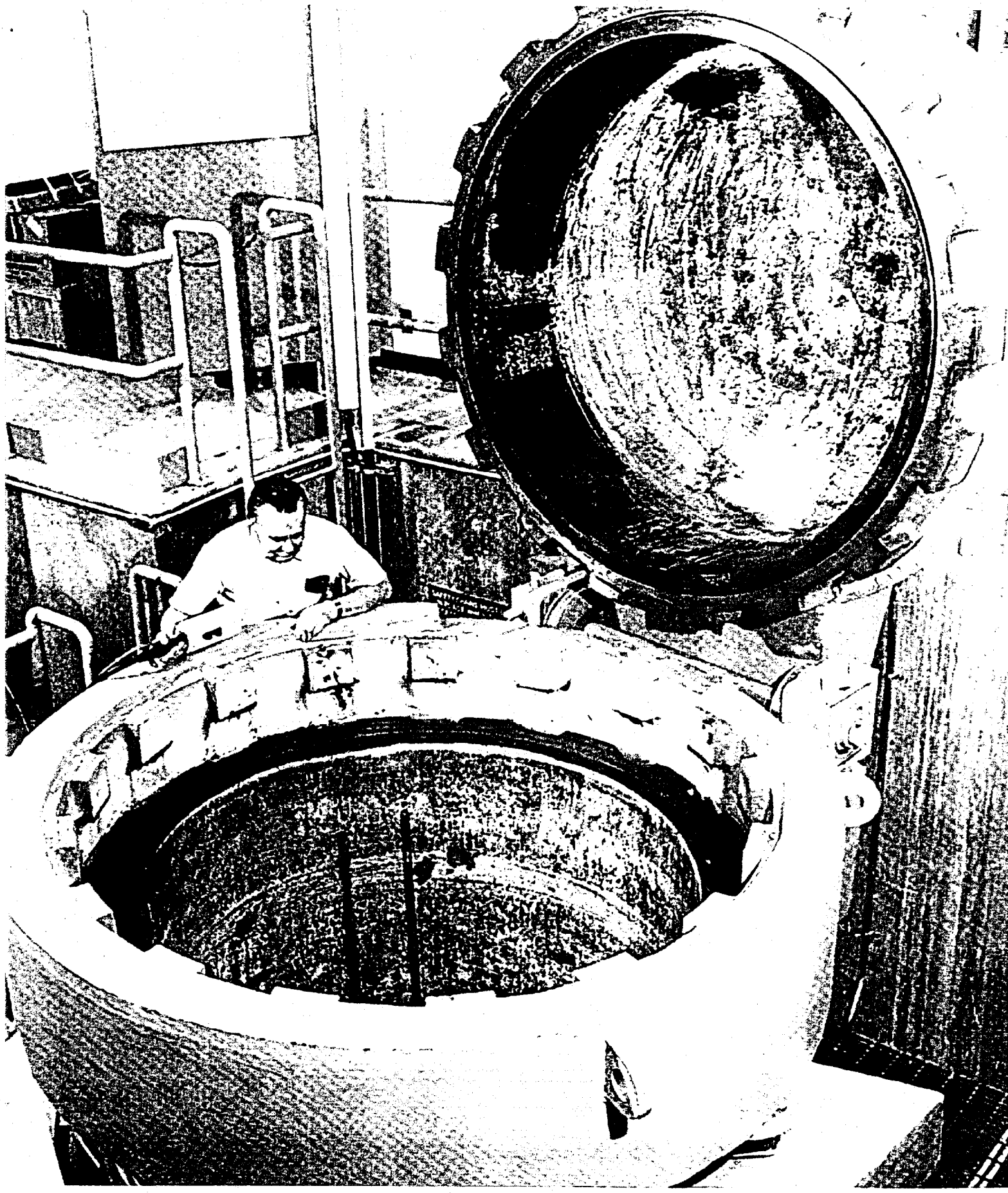
  —Spaces between fuel pins in CANDU[a] SNF are smaller than those in LWR SNF

[a]Canadian Deuterium Uranium reactor

ORNL DWG 97C-283



Full Scale Cross Section of CANDU and PWR Fuel Assemblies



Use of DUO_2 Fill In SNF Waste Packages Provides A Disposal Method For Excess Depleted Uranium

- The world inventory of excess depleted uranium is ~1 million tons
- The technology could beneficially use most of this depleted uranium

Conclusions

- The use of DUO_2 as a fill material in SNF waste packages may significantly improve repository performance
- Repository costs may be reduced by disposal of excess depleted uranium and providing a lower-cost method to assure repository performance
- Additional work is required to better quantify benefits and costs