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

PRESENTATION TO
THE NUCLEAR WASTE TECHNICAL REVIEW BOARD

SUBJECT:  SPENT FUEL CHARACTERIZATION OVERVIEW

PRESENTER:  RAY B. STOUT

PRESENTER'S TITLE AND ORGANIZATION:  TECHNICAL AREA LEADER
                                        WASTE FORM CHARACTERIZATION
                                        LAWRENCE LIVERMORE NATIONAL LABORATORY
                                        LIVERMORE, CALIFORNIA

PRESENTER'S TELEPHONE NUMBER:  (415) 423-3965

AUGUST 28-29, 1990
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SPENT FUEL (SF) OVERVIEW

OUTLINE

• INTRODUCTION

  • DISTRIBUTION ASPECTS OF PHYSICAL, CHEMICAL, AND RADIONUCLIDE PROPERTIES VERSUS BURNUP AND FISSION GAS RELEASE

  • CONCEPTUAL MODELS UNDER DEVELOPMENT TO PLAN TESTS AND TO DESCRIBE SF RESPONSES
WHY PERFORM SPENT FUEL CHARACTERIZATION?

OBJECTIVE OF SF CHARACTERIZATION ACTIVITIES:

TO PROVIDE DATA, TESTING, AND MODELS THAT DESCRIBE DEGRADATION AND RADIOACTIVE RELEASE RESPONSES OF SF FOR WASTE PACKAGE AND SYSTEM PERFORMANCE ASSESSMENTS IN THE YUCCA MOUNTAIN PROJECT
WHY PERFORM SPENT FUEL CHARACTERIZATION

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SPENT FUEL RESPONSE OVERVIEW

Potential event sequence in time

- Gaseous release response from cladding
- Cladding degradation response
- \( \text{UO}_2 \) oxidation response
- \( \text{UO}_2 \) dissolution response
- Geochemistry solution response

\( t \) time

Gas pressure

\( \text{O}_2 \)

Water
SPENT FUEL
CHARACTERIZATION OVERVIEW
(CONTINUED)

- PRELIMINARY WASTE FORM CHARACTERISTICS REPORT (MARCH 91)

- CONTENTS OF REPORT
  - PHYSICAL PROPERTY DATA FOR EXISTING AND PROJECTED SFWF INVENTORIES
  - RADIONUCLIDE DATA FOR EXISTING AND PROJECTED SFWF INVENTORIES
  - MODELS AND TEST DATA FOR SPENT FUEL DEGRADATION
NUCLEAR FUEL
INSERT
SPENT FUEL OVERVIEW

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ASSEMBLY CLASS QUANTITIES AND TYPICAL DIMENSIONS

GE BWR/4,5,6
GE BWR/2,3
Dresden 1
Humboldt Bay
Blg Rock Point
Lacrosse
Elk River
WE 15 x 15 array
WE 17 x 17 array
BW 15 x 15 array
CE 14 x 14 array
WE 14 x 14 array
CE 16 x 16 array
Haddam Neck
Pallsades
San Onofre 1
Fort Calhoun
Yankee Rowe
Saint Lucle 2
Indian Point 1
BW 17 x 17 array
South Texas 1&2

WE PWR typical attributes
Assembly length ~ 160 in.
Assembly area ~ 8.5 in. x 8.5 in.
Assembly weight ~ 1373 lbs.
#Rods/Assembly ~ 264
#Pellets/Rod ~ 288

GE BWR typical attributes
Assembly length ~ 175 in.
Assembly area ~ 6 in. x 6 in.
Assembly weight ~ 562 lbs.
#Rods/assemble ~ 63
#Pellets/Rod ~ 360

Dec. 31, 1987
2020 A.D.
SPENT FUEL INVENTORY-
HISTORY AND PROJECTION

BURNUP (GWd/MTU)

THOUSANDS OF TONS OF WASTE

BWR
PWR

1968-1987
1968-2037

CURRENT INVENTORY 20,000
PROJECTED INVENTORY 80,000

COURTESY: ORNL

SPFOVW5 A36/8-26/29-90  10
ILLUSTRATIVE ROD POPULATION DISTRIBUTION OF BURNUP

Histogram of burnup

Approximation to the histogram

Average

Quantity of spent fuel
# rods/(unit burnup)

Burnup, MWd/KgM

10 20 30 40 50 60 70 80
METHOD FOR CORRELATING MAXIMUM OXIDE THICKNESS WITH BURNUP

- CE (Andrews et al. 1988)
- B&W (Newman 1987)
- MCC (CE fuel rods)

Conservative correlation derived from available data

Maximum oxide thickness, μm

Burnup, MWd/kgM

0 10 20 30 40 50 60

106

104

104

104
ILLUSTRATIVE ROD POPULATION DISTRIBUTION OF MAXIMUM OXIDE THICKNESS

Maximum oxide distribution function

Average

Quantity of spent fuel
# rods/(unit oxide thickness)

Maximum oxide thickness, \( \mu m \)

0 25 50 75 100 100
ROD POPULATION DISTRIBUTION FOR OTHER ATTRIBUTES

SOME EXAMPLES:

- RODS PER UNIT CARBON-14 VERSUS CARBON-14 IN CLADDING
- RODS PER UNIT HYDROGEN VERSUS HYDROGEN IN CLADDING
- RODS PER UNIT DECAY HEAT VERSUS DECAY HEAT IN SPENT FUEL
- RODS PER UNIT RADIOACTIVE SPECIES VERSUS RADIOACTIVE SPECIES
ILLUSTRATIVE ROD POPULATION DISTRIBUTION OF FISSION GAS RELEASE

QUANTITY OF SPENT FUEL

#RODS/(UNIT FGR)

PERCENT FISSION GAS RELEASE (FGR)
METHOD OF CORRELATING GAP AND GRAIN BOUNDARY INVENTORY WITH ROD-AVERAGE FISSION GAS RELEASE

\[
\begin{align*}
\% \text{Total inventory plated out on cladding} & \quad \text{Conservative correlation based on U.S. and Canadian data}
\end{align*}
\]

\[
\begin{align*}
\text{% Fission gas release} & \\
\text{137 Cs} & \\
\text{129 I} & \\
\end{align*}
\]
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SPENT FUEL RESPONSE OVERVIEW

Potential event sequence in time

Gaseous release response from cladding
Cladding degradation response
UO₂ oxidation response
UO₂ dissolution response
Geochemistry solution response

HARRY SMITH (PNL)
HARRY SMITH (PNL)
ROBERT EINZIGER (PNL)
CHARLES WILSON (PNL)
HERMAN LEIDER (LLNL)
CAROL BRUTON (LLNL)
GASEOUS RELEASE (CARBON-14) FROM CLADDING

APPROXIMATELY 2% OF TOTAL CARBON-14 INVENTORY RELEASED RAPIDLY (~350°C) IN 8 HOURS

MODEL - DIFFUSION EQUATION
ZIRCALOY CLADDING DEGRADATION

Oxide film failure - stress corrosion cracking inhibitor

Zirconium oxide film

Oxide film is highly protective because of the large increase (~10%-15%) in volume (results in large compressive stress state in film) that occurs when zirconium transforms to zirconium oxide. Gas pressure - due to initial gas pressure of fuel rod plus fission gas released pressure.

Model - clad formation response due to pressure for elastic, plastic (creep), thermal and hydride platelet strains in the material.

Result: Gas pressure - temperature - hydride - time dependent failure response.

Thin-walled tube
Radius/thickness ~8
ZIRCALOY CLADDING DEGRADATION
(CONTINUED)

Hydrogen diffuses into the Zircaloy clad when the oxide film forms at reactor operating temperatures.

Hydrogen solubility limit in Zircaloy is ~160ppm at 325°C and decreases with temperature.

During cooling, hydrogen precipitates as brittle zirconium hydride platelets whose orientation depends on stress.

Circumferential planar platelets shown do not lead to degradation.

Radial planar platelets (perpendicular to ones shown) can lead to a crack propagation pathway through the cladding thickness.

Model - Hydride deformation coupled with hydride thermodynamic precipitation.

Result: stress dependent platelet orientation and failure response.
ZIRCALOY CLADDING DEGRADATION

(CONTINUED)

Zircaloy–fluoride corrosion response

Zirconium oxide film

ZIRCALOY HAS A PITTING CORROSION RESPONSE THAT IS LINEAR WITH RESPECT TO THE HYDROFLUORIC ACID CONCENTRATION. THE PITTING CORROSION REDUCES THE EFFECTIVE CLAD WALL THICKNESS WHICH RESULTS IN AN INCREASE IN STRESS. ALSO, PITTING COULD EVENTUALLY RESULT IN PINHOLE PATHWAYS THROUGH THE CLADDING.

MODEL - ELECTRO-CHEMICAL CORROSION

Thin-walled tube
Radius/thickness ~ 8
ZIRCALOY CLADDING DEGRADATION - EXPECTED TOTAL MODELING RESPONSE

SCHEMATIC OF CUMULATIVE RESPONSE CURVE FOR PROBABLE NUMBER OF FAILED SPENT FUEL RODS AT A TIME t

- Initial number rods failed
- High temperature high pressure failures
- Hydride stress reorientation failure
- Zircaloy-fluoride corrosion failures
SPENT FUEL OXIDATION RESPONSE

FUEL PELLETS, NOMINALLY 0.5cm TO 0.6cm RADIUS AND ~2cm LENGTH, FRACTURE INTO FRAGMENTS DUE TO THERMAL STRAINS DURING FIRST FULL POWER CYCLE.

FUEL FRAGMENTS OXIDIZE AFTER CLADDING BREACH.

MODEL - OXIDATION KINETICS DEPEND STATISTICALLY ON FRAGMENT SIZES AND SHAPES IN A TEST SAMPLE; ANY FRAGMENT CAN BE SUBDIVIDED INTO DIFFERENT SIZED PYRAMIDAL VOLUME SUBSETS TO OBTAIN A STATISTICAL DISTRIBUTION FUNCTION.
SPENT FUEL OXIDATION RESPONSE

(Continued)

RESULTS: TESTS ON SPENT FUEL FRAGMENTS HAVE SHOWN A GRAIN BOUNDARY OXIDATION FRONT MOVING INTO FRAGMENTS, FOLLOWED BY A SPATIAL ZONE WHERE OXIDATION OF INDIVIDUAL GRAIN VOLUMES OCCUR

Fractured pellets

Clad

Zone of grain volume oxidation

UO$_{2}$

Grain boundary oxidation front

Fragment cross-section
SPENT FUEL RADIONUCLIDE RELEASE

AQUEOUS RELEASE OCCURS FROM THE PELLET-CLADDING GAP, FROM CRACKS AND GRAIN BOUNDARIES, AND FROM FUEL GRAINS. RELEASE CAN DEPEND ON AREA, TEMPERATURE, BURNUP, SOLUBILITY, WATER CHEMISTRY, AND WATER FLOW RATE.

MODEL - THERMOCHEMICAL APPROACH TO DESCRIBE THE DISSOLUTION RATES AND RELEASE OF SOLUBLE SPECIES SUCH AS Cs, I, Tc, Sr, AND C, AND RELEASE OF THE SOLUBILITY LIMITED SPECIES SUCH AS THE ACTINIDES.
SPENT FUEL RADIONUCLIDE RELEASE
(CONTINUED)

RESULTS: "FLOWRATE" CONTROL AND SEMI-STATIC EXPERIMENTS PROVIDE INPUT TO MODELS WITH COUPLING TO EQ 3/6 GEOCHEMICAL SIMULATION
SPENT FUEL TESTING

- MECHANISTIC MODEL DEVELOPMENT BASED ON SHORT-TERM TESTING NECESSARY TO OBTAIN LONG TIME RESPONSE
- TESTING PERFORMED OVER A RANGE OF EXPERIMENTAL VALUES THAT EXCEED THE PREDICTED REPOSITORY CONDITIONS ON THE HIGH AND LOW SIDE WHENEVER POSSIBLE
- THIS MEANS THAT RESPONSE PREDICTIONS ON THE EXPERIMENTAL VARIABLES CAN BE MADE USING CONSERVATIVE INTERPOLATION RATHER THAN BY RISKY EXTRAPOLATION

![Graph showing response Y vs. test variables with repository conditions highlighted]