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Briefing for NRC Commissioners
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DOE Nuclear Waste Management Program at LLNL



- • **Past and present waste management R&D at LLNL**
- **Waste package licensing issues under 10 CFR 60**
- **An alternative strategic approach to high level waste management**

LLNL started in the high-level waste management program in 1976



- **Geochemical Modeling, 1977-present**
- **Nevada Project**
 - Spent Fuel Test-Climax, 1977-1985**
 - Waste Package for Tuff Repository, 1982-present**
- **Rock Mechanics and Geochemistry of a Salt Repository, 1976-1987**
- **SYNROC Project, 1979-1983**

Requirements for the engineered barrier system in 10 CFR 60



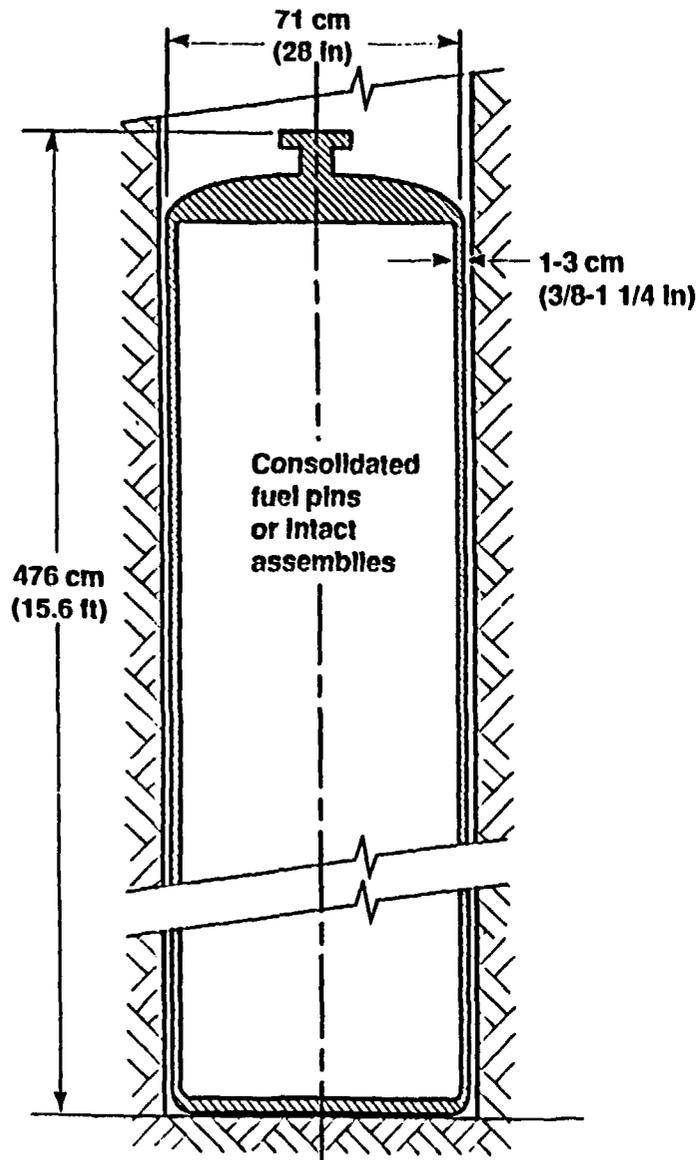
- 60.111** Requires up to 50 years retrievability of waste following start of emplacement.
- 60.113** Requires
 - **"Substantially complete containment" of the HLW within the waste packages for a period between 300 and 1000 years after repository closure.**
 - **Following the containment period, control release from the engineered barrier system to a rate not to exceed 1 part in 100,000 of the 1000-year inventory per nuclide per year.**
- 60.135** Sets general criteria for benign package-environment interaction, and specific criteria such as requiring solid waste forms and sealed containers.
- 60.21** Requires evaluation assuming "unanticipated processes and events", and a comparative evaluation of alternatives.

LLNL Yucca Mountain Project–Waste Package Activities

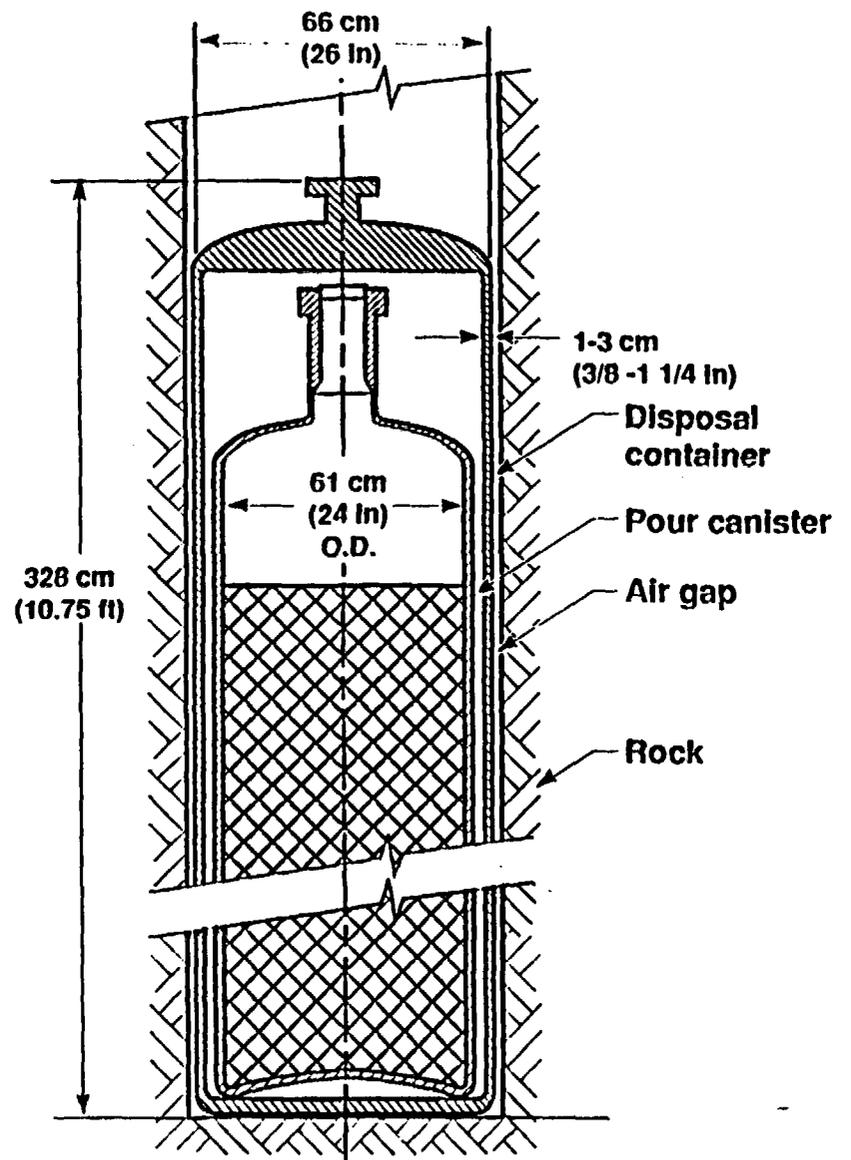


- **Service environment of the waste package**
- **Design integration and specification**
- **Laboratory materials testing**
 - **Waste form**
 - **Spent fuel**
 - **Glass**
 - **Metal barrier and alternate container materials**
 - **Other repository materials**
 - **Integrated testing of interaction of all materials**
- **Test of full-scale package**
- **Testing in field environment**
 - **environmental response**
 - **prototype package**
- **Assessment of package performance**

Two types of waste packages will be placed in the repository at Yucca Mountain



**Spent fuel containers
(25,000 to 35,000)**



**Waste glass
containers (~14,000)**

LLNL Technical Strategy



Without access to the Yucca Mountain repository horizon, design parameters must be assumed using best available information.

LLNL strategy follows a dual path prior to completion of site characterization at Yucca Mountain, preparing for both expected conditions (reference path) and a bounding case (alternative path).

Reference Path for Yucca Mountain Waste Package Strategy



Assumes that the expected benign waste package environment can be demonstrated during licensing:

- **based on a metal container**
- **uses currently available technology**
- **limits the amount of water contacting the waste package**
- **depends on factors in addition to the container:**
 - **an air gap around the container**
 - **the fuel cladding as a barrier**
 - **constraints on chemistry of water contacting packages**
 - **the insolubility of most fuel radionuclides**
 - **exemption for several high solubility nuclides with a very small inventory and insignificant public health risk**

Alternative Path for a Yucca Mountain Waste Package Strategy



Assumes that proving a benign environment in licensing will be sufficiently difficult so that the reference path would be imprudent

- **a more robust engineering solution**
- **uses material that is more geochemically stable than reference metals in the Yucca Mountain environment**
- **is at or slightly beyond present fabrication technology limits**
- **can withstand both more water contacting the packages and a more aggressive water chemistry**
- **has the potential to meet all performance requirements with the container alone**

Container Materials and Concepts



Reference Path

Austenitic

304L Fe₇₀ Cr₂₀ Ni₁₀

316L Fe₆₈ Cr₁₈ Ni₁₂ Mo₂

825 Fe₃₀ Cr₂₂ Ni₄₂ Mo₃ Ti₁ Cu₂

Copper Based

CDA 102 Cu_{99.95}

CDA 613 Cu₉₂ Al₆ Fe₂

CDA 715 Cu₇₀ Ni₃₀

Alternate Path

Ceramics

Graphite

Bi Metals

Other Single Metals

Coatings

Fillers

Thicker wall metals

DOE Nuclear Waste Management Program at LLNL



- **Past and present waste management R&D at LLNL**
- • **Waste package licensing issues under 10 CFR 60**
- **An alternative strategic approach to high level waste management**

Based on what we know today, these items either will be very contentious in licensing a Yucca Mountain Repository or lead to unnecessary effort and expense



- **Interpretation of key phrases in 10 CFR 60**
 - **Substantially complete containment**
 - **Boundary of the engineered barrier system**
 - **Anticipated processes and events**

- **Certain explicit requirements of 10 CFR 60**
 - **Release limit evaluated on an annual basis**
 - **Release limit same for all radionuclides**
 - **NRC controls radionuclides that EPA exempts!**

For the spent fuel waste form, technical information available today shows the radionuclides for which licensing challenges can be expected



- **Gaseous radionuclides**
 - C-14, Kr-85, and H-3

- **Highly soluble radionuclides**
 - Sr-90 and Cs-137 during containment

 - Tc-99, Cs-135, I-129, C-14, and Mo-93 during isolation

The fundamental insolubility of most radionuclides in the spent fuel inventory provides assurance of public safety



- **Am, Pu, Np, and U comprise 98.4% of the inventory at 1000 years.**
- **A few gaseous and highly soluble radionuclides that are of limited public health concern are driving the DOE licensing strategy.**
- **The cost and benefit appear to be out of proportion for these few radionuclides.**

Issues posed by gaseous radionuclides



- **Gaseous radionuclides can cross the EBS boundary even in absence of liquid water**
- **Only three radionuclides to consider - C-14, Kr-85, & H-3**
- **EPA standard recognizes insignificance and does not control Kr-85 & H-3 release to accessible environment (half-life <20 yr) - but NRC controls at EBS boundary!**
- **Major strategy driver is C-14 on exterior of fuel cladding because limits are 0.1 Ci (containment) and 1 Ci (isolation) per year. One power plant releases ~ 10 Ci per year!**
- **Based on estimated (poorly known) inventory of C-14, container failures (assuming 30,000 spent fuel containers) are limited to:**
 - **No to 3 containers in any year during containment period (1,000 yr)**
 - **30 containers in any year in isolation period (10,000 yr)**

Issues posed by readily soluble radionuclides



- **Some scenarios show short duration "spike" releases of a few radionuclides can occur from fuel in failed containers with limited liquid water**

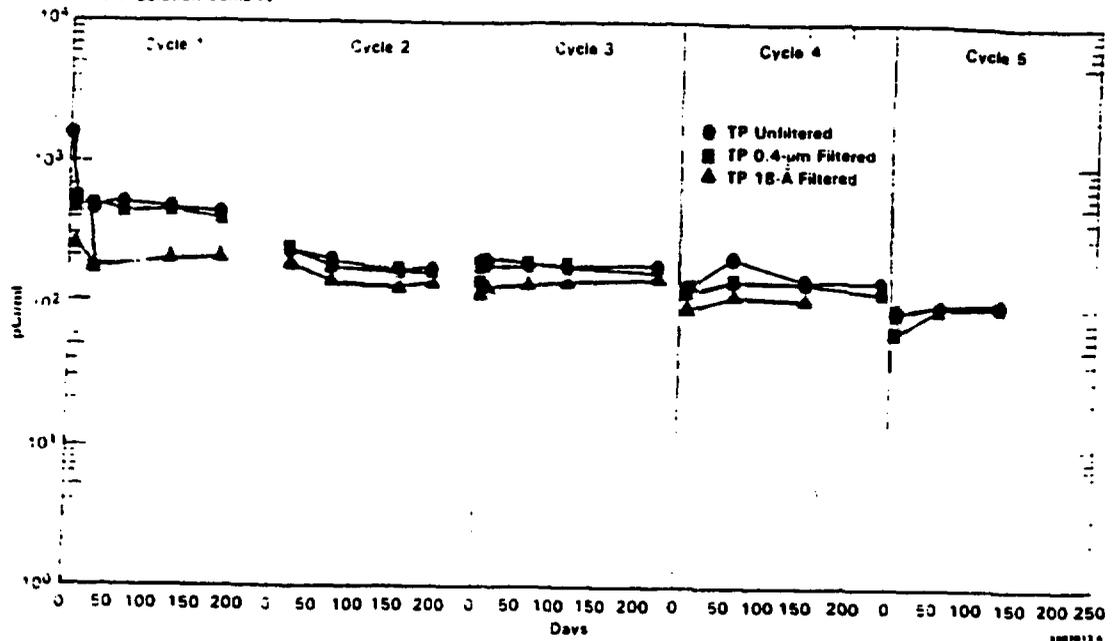
- **A few radionuclides are readily available in "gap-grain boundary" inventory**
 - **as much as 2% of total inventory in unoxidized fuel**
 - **may be more in oxidized fuel - not much data yet**

- **Radionuclides of concern:**
 - **Sr-90 and Cs-137 during containment - 30-year half-life, initial large inventory**
 - **Tc-99, Cs-135, I-129, C-14, and Mo-93 during isolation**

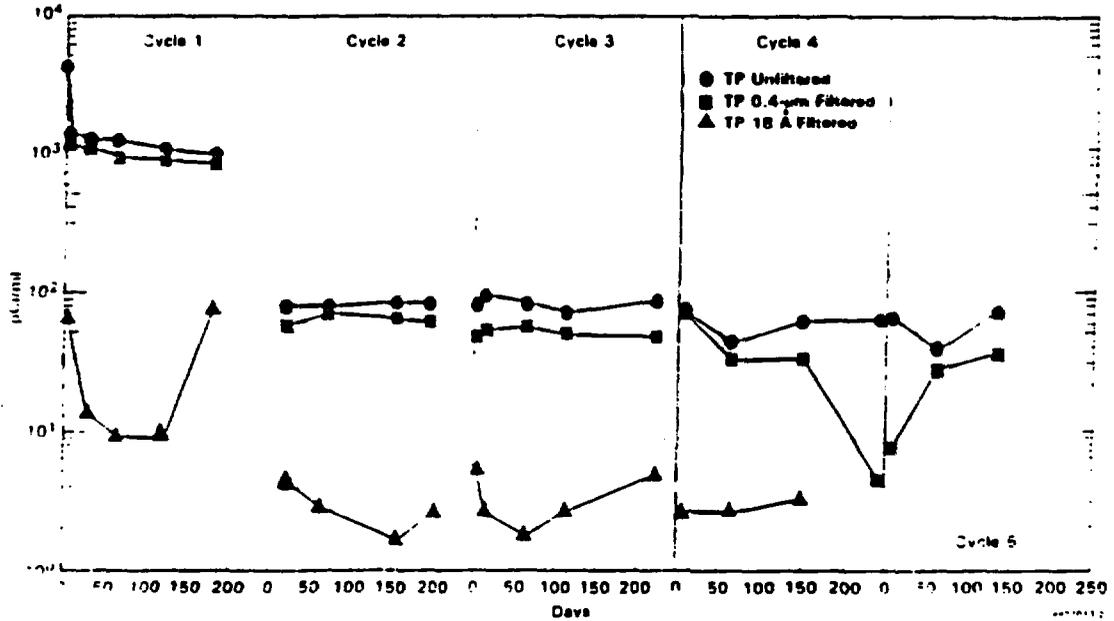
PWR Spent Fuel Inventory at 1,000-years. (33GWd/MTU)

Radionuclide	% activity	% Activity for element	Cumulative % activity
²⁴¹ Am	51.32	52.23	52.32
²⁴³ Am	0.90	"	
^{242+242m} Am	0.00	"	
²⁴⁰ Pu	27.47	45.11	97.34
²³⁹ Pu	17.53	"	
²⁴¹ Pu	0.008	"	
²⁴² Pu	0.099	"	
²³⁷ Np	0.057	0.954	98.29
²³⁹ Np	0.897	"	
⁹⁹ Tc	0.747	0.747	99.04
⁵⁹ Ni	0.294	0.316	99.35
⁶³ Ni	0.022	"	
²³⁴ U	0.117	0.152	99.19
²³⁵ U	0.001	"	
²³⁶ U	0.016	"	
²³⁸ U	0.018	"	
^{93m} Nb	0.105	0.176	99.36
⁹⁴ Nb	0.071	"	
⁹³ Zr	0.111	0.111	99.48
¹⁴ C	0.079	0.079	99.55
²³³ Pa	0.057	0.075	99.63
^{234m} Pa	0.018	"	
^{126+126m} Sb	0.050	0.050	99.68
¹²⁶ Sn	0.044	0.044	99.72
⁷⁹ Se	0.023	0.023	99.75
¹³⁵ Cs	0.020	0.020	99.77
²³⁴ Th	0.018	0.019	99.79
²³¹ Th	0.001	"	
¹⁵¹ Sm	0.009	0.009	99.80

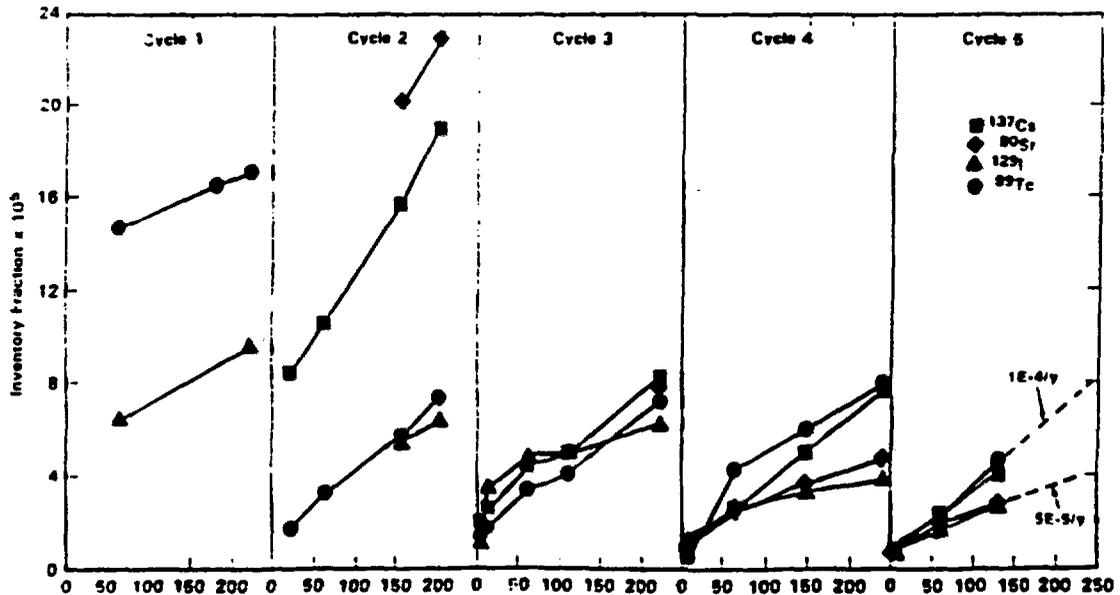
^{239, 240}Pu in Solution Samples



²⁴¹Am in Solution Samples



Fission Products in Solution -- HBR-2-25 Test



Other concerns with demonstrating meeting the performance objectives



- **Same numerical requirement for all radionuclides whereas limits in 40 CFR 191, Table 1, vary over 3 orders of magnitude depending on the specific radionuclide**
- **Evaluation period is annual, whereas 40 CFR 191 limit is cumulative**
- **Interpretation of where release is to be measured**
- **Possible inclusion of unlikely processes and events as "anticipated"**

Same numerical requirement for all radionuclides



PROBLEM: Though 40 CFR 191 allows release limits to vary over 3 orders of magnitude depending on the specific nuclide, 10 CFR 60 sets an invariant limit to be demonstrated on a per nuclide basis.

- **In 40 CFR 191, Cs-135 is allowed 10 times the release of actinides**
- **In 40 CFR 191, Tc-99 is allowed 100 times the release of actinides**

Table 1 in 40 CFR 191

**RELEASE LIMITS FOR CONTAINMENT REQUIREMENTS
(Cumulative Releases to the Accessible Environment
for 10,000 Years After Disposal)**

<u>Radionuclide</u>	<u>Release Limit per 1000 MTHM or Other Unit of Waste (curies)</u>
Americium-241 or -243	100
Carbon-14	100
Cesium-135 or -137	1000
Iodine-129	100
Neptunium-237	100
Plutonium-238, -239, -240, or -242	100
Radium-226	100
Strontium-90	1000
Technetium-99	10000
Thorium-230 or -232	10
Tin-126	1000
Uranium-233, -234, -235, -236, or -238	100
Any other alpha-emitting radionuclide with a half-life greater than 20 years	100
Any other radionuclide with a half-life greater than 20 years that does not emit alpha particles	1000

Evaluation period is annual



PROBLEM: Determination on a per year basis rather than moving average or cumulative total basis

- **It is virtually impossible to demonstrate that a "spike release" will not occur.**
- **At the edge of the disturbed zone, a spike release will have been smoothed by physical dispersion and other processes, so that a yearly accounting will become a meaningless point of contention.**
- **A better phrase would be "one part in 100,000 rate per year, using a 10 (or 100) year moving average".**

Interpretation of where release is to be measured



PROBLEM: Rock within the disturbed zone not included as part of the engineered barrier system--borehole wall is boundary of EBS

- **Spike release in any given year would not violate the annual limit if the rock out to 50 meters (the disturbed zone) were included in the EBS definition.**
- **Even a few meters of rock might allow for enough dispersion to reduce spike release to annual limit.**
- **Because retardation due to exchange, dispersion, dilution, sorption, reaction, and other processes is used in determining compliance with the total system performance objective outside the disturbed zone, these processes should be considered within the disturbed zone.**

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Potential opportunities to improve chances of demonstrating compliance with the regulations for high-level waste



- **Conservative nature of regulations and scattered unreasonable requirements.**
- **Unprecedented regulatory time-span with no agreed methodology for prediction.**
- **Technical uncertainty from unvalidated models and lack of site specific in-situ data.**

Technical uncertainty can be reduced by placing high level waste in the ultimate repository for several decades



- **Needed data are from the repository horizon.**
- **The model uncertainties are greatest at full scale and long times.**
- **Retrievable emplacement of waste is the best source of heat and radiation.**

There are a variety of ways to achieve retrievable emplacement of waste



- **Conceptually the simplest is to construct a full-scale underground MRS at Yucca Mountain that could be converted to a repository in 50 years.**
- **A more step-wise approach would emplace increasingly larger increments of waste starting with very small demonstration panels and leading to licensed modules of a repository at the Yucca Mountain site.**
- **A variation would have the demonstration emplacements be nearby, but outside the repository block.**

Early Access to the Repository Horizon is Vital



- **Physical access will rapidly resolve some uncertainties**
 - **Water flux through horizon**
 - **Details of fracture system**

- **Some uncertainties are resolved only with full scale, long-term testing over a representative volume of the repository**
 - **Details of rock response to heat sources**
 - **Validation of models**

- **Some uncertainties will not be addressed by currently planned access**
 - **Spatial variability**
 - **Radiation effects**

Concepts proposed for OCRWM strategy



Refocus the near-term priority of the National Program on DOE acceptance of waste rather than permanent disposal.

Carry out early demonstrations of the technical feasibility of disposal in unsaturated tuff.

Work with NRC to establish phased licensing of a repository with the objective of near-term licensed disposal of a portion of the waste inventory.

Take advantage of shift from emphasis on early disposal to benefit from technological advances of next several decades.

Refocus the near-term priority of the national program on DOE acceptance of waste rather than permanent disposal



- **There is no technical nor safety need for early disposal.**
- **Conversely, doing it right rather than fast should be priority--perception has been that schedule is more important than safety.**
- **U.S. is alone among nuclear nations in focus on early disposal.**

Carry out early demonstrations of the technical feasibility of disposal in unsaturated tuff



- **Permanent disposal is necessary to close the fuel cycle.**
- **MRS will fail like RSSF concept if public perceives that temporary storage results in doing nothing on permanent disposal.**
- **Needed to meet legal requirements (California law, Waste Confidence Rulemaking).**
- **A repository is a first-of-a-kind facility--good engineering practice calls for demonstrating technology step by step.**

Work with NRC to establish phased licensing of a repository with the objective of near-term licensed disposal of a portion of the waste inventory



- **Equivalent to low power license for a nuclear power plant**
- **Logical next step after demonstration stage**
- **50-year retrievability makes this an easy conceptual evolution of the present single 70,000 metric ton step**
- **Legal and public perceptual needs can be satisfied by 1,000 or 5,000 metric ton repository--70,000 metric tons not needed now.**

Take advantage of shift from emphasis on early disposal to benefit from technological advances of next several decades



- **Advanced container materials that have improved performance and are cost effective**
- **Advances in site characterization and predictive modeling technology**

LICENSING ISSUES POSED BY GASEOUS RADIONUCLIDES IN SPENT FUEL

In the expected unsaturated dry environment of the Yucca Mountain site, a breached container could release only gaseous radionuclides. These gases, tritium (12.3 yr half-life), Kr-85 (10.7 yr half-life), and C-14 (5730 yr half-life), have a potential path to the surface above the repository via gaseous diffusion and advection. Only C-14 is an issue post-closure, because 40 CFR 191 does not control radionuclides with half-lives less than 20 years. Although C-14 poses a negligible threat to public health and safety if released in small amounts in a sealed repository, it is a significant factor driving the waste package design, because it might technically violate the NRC regulations for substantially complete containment or release control.

There are several sources of gaseous radionuclides in spent reactor fuel: the exterior of fuel cladding, the rod plenum and gap between the cladding and the fuel matrix, the UO₂ matrix itself, and the bulk of the cladding and other hardware.

Cladding exterior. Release of C-14 from the cladding exterior is dominantly as CO₂ and is controlled by a thermally activated diffusion process in the presence of oxygen. The inventory of this C-14 is somewhat uncertain as it has three primary sources: neutron bombardment of nitrogen impurities within the zircaloy cladding, neutron bombardment of oxygen that has been incorporated in the oxide layer on the cladding, and possibly deposition from the reactor cooling water on the cladding as "crud" and oxide film. However, it is estimated to be about 20 percent of the total C-14 inventory of PWR spent fuel. Recent scoping experiments have shown that ~ 10% of that inventory (2 % of the total inventory) of C-14 is released as CO₂ after 8 hours exposure to air at 350 degrees C. A much smaller though significant amount (slightly less than 0.1 % of the total inventory) is released under the same conditions in Ar.

Plenum. The plenum gas is readily available upon breach of the cladding, which could occur prior to breach of the container. Because it consists mostly of short half-lived Kr-85 and tritium, it is of concern only in the pre-closure phase of the repository.

Fuel matrix. Gaseous release from the fuel matrix is controlled by the rate of matrix oxidation or dissolution, which can occur only after

both the container and cladding are breached. The only gaseous radionuclide for which this source mechanism is significant is C-14.

Because 10 CFR 60 was written prior to quantitative studies of radionuclide release from spent fuel, it did not consider the rapid release fraction of C-14 that is on the external surface of irradiated fuel cladding. Furthermore, the regulations were written based on analysis of a saturated site. In an unsaturated site, a small amount of C-14 can escape from a few breached containers and, by passing the boundary of the emplacement hole wall, place the repository in technical violation of either the containment or the controlled release performance objective.

In a repository with 30,000 containers of spent fuel, breach of only 15 in any one year between 1000 and 10,000 years post-closure might constitute a technical violation of the controlled release performance objective, yet release only ~ 1 Ci of C-14. Under the current working definition of substantially complete containment, breach of only two containers in any year up to 1000 years post-closure might be a technical violation, yet release only ~ 0.1 Ci. By contrast, a PWR nuclear power plant releases about 6 to 12 Ci/yr to the environment, and a BWR plant about 12 to 14 Ci/yr.

There is no explicit requirement in 10 CFR 60 for DOE to include post-closure controls for tritium and krypton-85 in the Site Characterization Plan, but there is also no explicit exemption. As noted above, 40 CFR 191 does not control radionuclides with half-lives less than 20 years. For C-14, both 40 CFR 191 and 10 CFR 60 are inconsistent with significantly larger releases allowed for operating reactors and reprocessing plants as well as the order of magnitude higher level of risk allowed in other EPA regulations for non-nuclear hazardous materials. Even the operational phase of the repository allows for higher release of gaseous radionuclides than the post-closure period.

These gaseous radionuclides pose no threat to public health or safety, yet are driving the waste package design. Early addressing of this licensing issue would allow resources to be focussed on more substantive issues.

Reference: R.A. Van Konynenburg, Review and Position Paper on Carbon-14 Release from the Proposed High-Level Nuclear Waste

Repository at Yucca Mountain, Nevada, Draft UCRL report, Lawrence
Livermore National Laboratory, September, 1989

LICENSING ISSUES POSED BY READILY SOLUBLE RADIONUCLIDES IN SPENT REACTOR FUEL

In early planning for disposal of waste from nuclear reactors, it was expected that the spent fuel would be reprocessed to remove fissile isotopes and potentially useful fission products, so that the high level waste would consist mostly of fission and activation products. Immobilization as a solid waste form, probably borosilicate glass, was expected. The direct disposal of spent reactor fuel was not considered seriously until the late 1970's. Experimental data on radionuclide release from actual spent fuel only became available in the late 1980's.

Standards in regulations must be achievable. In drafting the current regulations for disposal of high-level waste (40 CFR 191 and 10 CFR 60), assumptions about achievability were based primarily on information available from dissolution testing of trial formulations of borosilicate-glass waste forms. Available evidence indicated that glass could meet a one part in 100,000 standard based on measured dissolution rates.

If the dissolution reactions involving solid waste forms are congruent then, by definition, all components of the waste are dissolved from the matrix at the same rate. The dissolution of borosilicate glass can be shown to be congruent, although the actual release is incongruent due to reprecipitation of less soluble components. For glass, the maximum release rate of any component is limited by the overall dissolution rate, which is controlled mostly by the solubility of Si in ground water. In this case, setting regulations limiting the release rate to "one part in 100,000 per year" for each radionuclide seems reasonable, because all radionuclides have the same dissolution rate and the maximum release rate can be determined readily.

For incongruent release, individual components have differing release rates, and the determination of maximum release rate is complex. Maximum release rates from spent fuel are not controlled by the congruent dissolution rates for the UO₂ matrix, as explained below. Because assumptions underlying the current regulations were based on a model of congruent dissolution, there is possible difficulty in demonstrating that the standard is achieved.

Reactor fuel consists of UO_2 fuel pellets encapsulated in Zircaloy tubes (cladding) that are sealed. During irradiation, certain fission products migrate first to grain boundaries and thermally induced fracture surfaces within the pellets, and then to the gap between the pellets and the cladding. This is called the "gap-grain boundary inventory". Not only are these radionuclides physically more accessible, but certain of them are highly soluble and can build up in any water in contact with the waste once the cladding is breached. These include isotopes significant during the containment period such as Sr-90 and Cs-137, and also long-lived fission products such as Tc-99, I-129, and Cs-135.

Dissolution of the gap-grain boundary inventory is the simplest and most likely path of aqueous radionuclide release from spent fuel in a repository. It would occur at any site regardless of the geology. Several site-specific mechanisms also have potential for incongruent release.

In a fractured, unsaturated environment such as Yucca Mountain, oxidation of the fuel by contact with air can occur prior to its being contacted by water. As a result of this oxidation, fission products previously locked in the UO_2 matrix are made available for selective leaching when later contacted by water.

Another mechanism is related to the presence of oxidizing groundwater. Although the dissolution of the UO_2 fuel matrix may be congruent, radionuclide release is incongruent because of subsequent precipitation of uranium and other actinides. If the fuel is immersed in oxidizing water, oxidative dissolution of the UO_2 fuel matrix will take place. Even though the water may be saturated with respect to other uranium-bearing phases, UO_2 is soluble and continues to dissolve. The dissolved uranium oxidizes and then precipitates as hexavalent uranium compounds. Other actinides also precipitate, but the soluble fission products remain in solution and are either transported away or continue to increase in concentration.

Thus, there exist three mechanisms for obtaining pulse or spike aqueous releases of readily soluble radionuclides from individual containers of spent fuel:

- (1) selective leaching of the gap-grain boundary inventory, which is approximately equal to the fission gas release of the fuel (< 2 % of the inventory);

- (2) selective leaching of oxidized fuel, which theoretically could expose nearly the entire affected inventory of fission products to rapid availability; and
- (3) build-up through essentially unlimited congruent dissolution of the UO_2 matrix as a result of re-precipitation of higher valence uranium.

Taken together, the above mechanisms suggest the likelihood of a spike release that might technically violate the 10 CFR 60 requirement that release be controlled for each radionuclide with an annual limit at the EBS boundary.

In most cases, such a release would have little relation to public health and safety or to meeting 40 CFR 191. The cited radionuclides make up a very small proportion of the inventory and the spike release will be offset by other years of lower release. Furthermore, Sr and Cs are readily sorbed by the rock, and Sr-90 and Cs-137 have short (~30 yr) half-lives. The exception to these statements is Tc-99, which is the third most abundant isotope at 10,000 years, making up 2.7% of the total activity. Tc-99 is also highly soluble and does not sorb to rock under oxidizing groundwater conditions.

Laboratory tests with unoxidized spent fuel have resulted in buildup to 0.1% of the Tc-99 inventory in less than one year (Wilson and Bruton, 1989, Fig. 4, p 8). Tests with oxidized spent fuel have resulted in Tc-99 buildup to 1% levels in less than one year (Wilson and Gray, 1990, Fig.6, p 9). Such results indicate that in a "bathtub" scenario (filling of a container with water that resides for years before draining rapidly), buildup of Tc-99 to a level of one percent or more of the inventory within a single container can occur within several years. At the one percent level, draining of water from only 30 such containers in any year (assuming 30,000 in the repository) would constitute a technical violation (cross the EBS boundary at the borehole wall) of the one part in 100,000 release limit for the total repository.

Although 10 CFR 60 has the same limit for all radionuclides, the 40 CFR 191 release limits vary over 3 orders of magnitude. Of particular interest is the fact that the limit for Tc-99 is 100 times greater than for the actinides, while Sr-90, Cs-135, and Cs-137 have a ten times greater limit than the actinides. There is an apparent recognition of this fact in 10 CFR 60.113b, wherein it is stated " On a case-by-case basis, the Commission may approve or specify some

other radionuclide release rate...provided that the overall system performance objective...is satisfied."

If site conditions at Yucca Mountain are shown to be as expected, radionuclide release will be insignificant even given the high solubility of the gap-grain boundary inventory. However, for certain unlikely but possible scenarios, one can postulate releases that technically violate current interpretations of 10 CFR 60. Because it will be years until conditions at the site can be verified, expensive engineering alternatives have been suggested to assure meeting the rule. However, consideration of other radionuclide release rates as allowed in 60.113b has not been invoked and it should. It is the simplest, most cost-effective approach to addressing the now-apparent and significant differences between spent fuel and glass as waste forms.

References

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