ACCEPTANCE CRITERIA CONSIDERATIONS FOR MISCELLANEOUS WASTES

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ABSTRACT

Extensive preparations are under way for the geologic disposal of conventional high-level wastes and spent fuels. However, the nonconventional wastes, whose volume is only a small fraction of that of conventional high-level wastes and which have greater activity levels than are permitted for shallow-land burial, may be routed to a geologic repository. This paper describes the latter wastes and presents two approaches whereby their disposal costs might be minimized while satisfying the intent of the regulations. The approaches are: (1) elimination of the 300- to 1000-y-life container, which might be justified in the absence of a significant thermal pulse (achievable by predisposal decay and/or dispersal in the repository), and (2) employment of a more effective migration barrier around the waste form to compensate for conservatively estimated, as opposed to conclusively determined, performance of the waste form as a barrier to the release of radioactivity.

INTRODUCTION

The primary radioactive wastes for geologic disposal are light-water reactor (LWR) spent fuels and vitrified high-level wastes (HLW). In addition to these, there are a number of other highly radioactive wastes (called miscellaneous wastes), most or all of which may be emplaced in a geologic repository. These include (1) special nuclear fuels, (2) metals activated by neutron irradiation, (3) commercial transuranic wastes, and (4) uranium oxide solids containing U-233. These wastes are small compared with main-line wastes in terms of total radioactivity, mass, and volume.

Applicable federal regulations (1 and 2) and preliminary nuclear waste acceptance criteria (3 and 4) for deep geologic disposal of radioactive wastes were formulated with LWR spent fuel and vitrified high-level wastes as the central consideration. Because of the great difference in quantities of the miscellaneous wastes (MW) as compared with the quantities of LWR spent fuels and vitrified HLW, it seems unreasonable to expend the same amount of resources on the development of both waste acceptance criteria and modified and/or characterized (as to release rate) waste forms for the small-volume, relatively low-activity-content MW as has been (and is being) expended on the main-line wastes. Although waste quantities may be small, the objective must be to satisfy the intent of the regulations while at the same time minimizing costs. An approach to accomplishment of this dual objective that is suggested in this paper involves conservative design of migration barriers located around the waste form as a technique to compensate for uncertainty of the waste form performance.

The NRC regulations (1) pertaining to wastes emplaced in a deep geologic repository (10 CFR 60) seek to ensure environmental protection by stipulating certain characteristics and performance levels for the waste package and for the geologic repository. The principal waste package requirements are retrievability, substantially complete containment during the period of high heat generation, and a specified maximum release rate from the waste package when heat release is no longer significant. No specific method is indicated for satisfaction of the requirements; therefore, it must be assumed that any method, or combination of methods, which can be shown to satisfy the requirements will be acceptable.

This paper will discuss some potential means for satisfying the intent of the applicable regulations pertaining to the effectiveness of the waste package for limiting the release of radionuclides in both the short and long terms. The objective of this paper is to explore the singular characteristics of certain MW and to identify some potential means by which the regulations relative to waste package performance might be satisfied in light of those characteristics. This exploration will include an examination of the requirements of the regulations and of the characteristics of the wastes, followed by a discussion of the available options.

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REGULATORY REQUIREMENTS

The Nuclear Regulatory Commission's requirements for "Disposal of High-Level Radioactive Wastes in Geologic Repositories" are set forth in 10 CFR 60 (1). Paragraphs 113 and 135 provide criteria for barriers and for the waste form. The major elements of these two paragraphs stipulate the following:

- Substantially complete containment within the waste package for a period of 300 to 1000 y, the actual time period being determined by the NRC.
- A maximum allowable release rate for any radionuclide from the engineered barrier system of 1 part in 100,000 per year of the amount remaining after a 1000-y decay period, except that the minimum required annual release rate for a radionuclide shall not be less than 10^{-8} of the total activity remaining after 1000-y decay.
- On a case-by-case basis the Commission may approve or specify variations in the above requirements provided that the overall system performance objective is satisfied.
- No explosives, pyrophorics, or chemically reactive materials in amounts sufficient to compromise the intended function of the repository.
- Solid waste form only (no liquids), with limits on particulates and combustibles.

Title 40, CFR 191 (2) "Environmental Radiation Protection Standards for Management and Disposal of Spent Nuclear Fuel, High-Level and Transuranic Radioactive Waste," stipulates a performance standard for the repository but does not delineate characteristics or performance of the waste package or any other individual portion of the radioactive waste confinement system.

The present paper is concerned primarily with meeting the containment and release rate standards of 10 CFR 60.113. The other elements of 10 CFR 60 are viewed as constituting relatively minor problems whose solutions are straightforward. It should be noted that the regulation provides NRC with the option of adjusting the complete containment and release rate standards, provided system objectives are satisfied.

OPTIONAL ROUTES TO COMPLIANCE WITH THE SHORT- AND LONG-TERM RADIONUCLIDE CONFINEMENT REQUIREMENTS OF 10 CFR 60

Because different waste types have divergent characteristics, it is apparent that identical treatment to ensure protection of the environment may be inappropriate. The generic operations potentially associated with the preparation of wastes for disposal and the options for waste package design are outlined in Fig. 1. The optional pathways for the several operations are also indicated. An exhaustive characterization of the vitrified HLW and LWR spent fuels is under way which will provide an accurate indication of release rates from these two waste forms; however, the experimental determination of waste form characteristics/performance is very expensive. Substantial expenditures can be justified for large volumes of waste such as those represented by LWR fuel and defense HLW; however, this might not be the case for the low-volume MW. If conservative assumptions were made for the performance of MW and a conservatively thick migration barrier were employed, it follows that the extent of waste form testing might be substantially reduced.

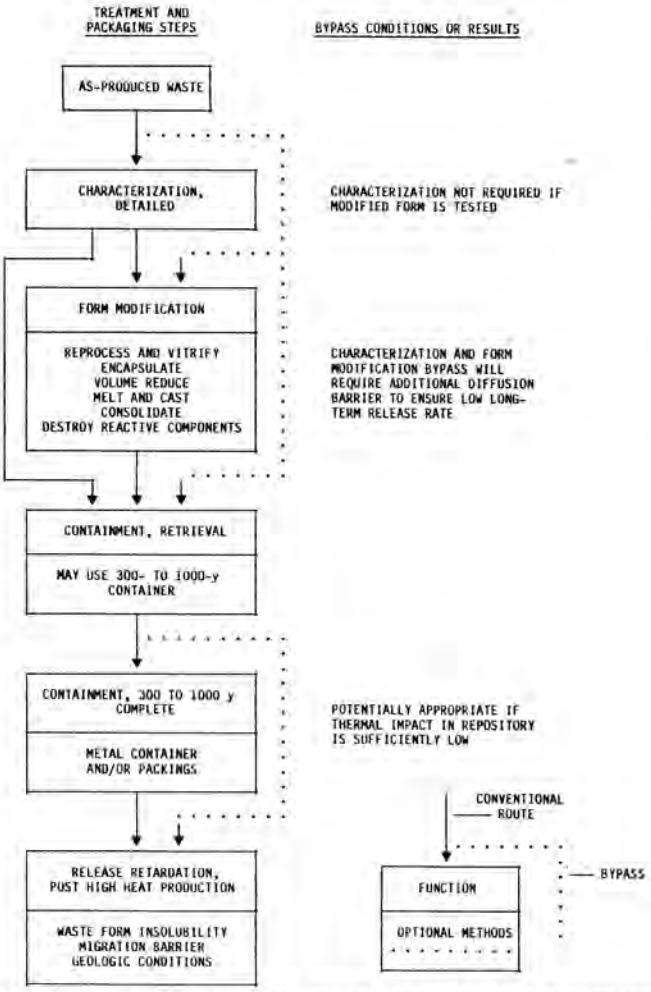

Fig. 1. Generic steps in waste treatment and packaging, with optional routings and methods of accomplishment shown.

Decisions concerning the thermal characteristics of the waste package and the areal heat load in the repository can affect the choice of options for 1000-y containment. For example, if the areal heat load is insignificant, there will be no period of effective high heat production; and if the low-heat-producing wastes are isolated from high-heat-producing wastes, there would appear to be no basis for a complete containment period. Significant precedent has been established for this perspective. In the United States, the unlicensed defense Waste Isolation Pilot Plant facility for disposal of transuranic waste does not use long-life waste containers because the waste does not produce a thermal pulse (5). Similarly in West Germany, the licensed, operating, 700-m-deep salt mine geological repository at Herfa Neurode (6) for heavy-metal wastes does not require long-life containers. Again, the basis is the lack of a thermal pulse. For minimal quantities of waste that have not been fully characterized, it might be appropriate to limit the areal waste density in the repository to such an extent that the heat generation from radionuclide decay would cause no significant increase in temperature within the repository. Under this condition, elimination of the "1000-y container" could apparently be justified. Wastes not housed within a 1000-y container would be emplaced in a repository zone which is widely separated from zones where high heat generating wastes are emplaced.

The conventional method of achieving substantially complete containment during the high-heat-production period is through use of a metal container, possibly in combination with a minimal migration barrier. It is conceivable that the containment objective could be achieved by using a thicker migration barrier. In this case, the container requirement might be limited to that needed for retrieval.

Long-term release retardation within a given geology is determined by the combination of waste form insolubility and the effectiveness of barriers for impeding movement of water and ionic materials. In the absence of adequate assurance that the combination of waste form and geology provides adequate release retardation, the use of (additional) migration barrier material around the waste form can serve to reduce the release rate (7-12). Thus, a migration barrier is potentially applicable to compensate for real or potential deficiencies in waste form performance. Determinations will be required for both waste form solubility and transport rate for water, ionic, and possibly colloidal materials through migration barriers.

MISCELLANEOUS SPENT FUELS

A number of miscellaneous spent fuels are currently in storage at various sites throughout the United States. Some of their characteristics are presented in Table I. These spent fuels can be divided into the following classes:

- High Temperature Gas-Cooled Reactor (HTGR)
- UO_2-ThO_2
- U-Th metal, NaK bonded
- UO_2-Zr clad
- Research Reactor - TRIGA
- Test Reactor - Molten Salt Reactor Experiment (MSRE)

Most of the fuels listed in Table I are relatively uncharacterized with respect to release rate in a repository. The heat generation quantities for most classes are small (by LWR fuel standards); and when taken in aggregate, the indicated heat generation from existing miscellaneous spent fuel is still quite low (<35 kW). Thus, it is conceivable that all of these wastes could be so packaged and distributed within an isolated portion of a geologic repository that they would produce no significant thermal effect. Further, the absence of credible data on waste form release characteristics, along with the small quantity and varied nature of the wastes, creates an incentive for providing a conservative thickness of migration barrier (in lieu of heavy dependence on a low waste form release rate) to ensure slow radionuclide release from the waste package. Many of the wastes have volumetric heat production rates at 10 y decay which are less than those of 1000-y-decayed LWR fuel irradiated to 33 GWd/Mg. Presumably, wastes exhibiting such low volumetric heat release rates would not have a significant thermal impact on a repository if placed in the repository at the same density as LWR fuel canisters. Those fuels having higher volumetric heat generation rates could be emplaced in small batches and similarly dispersed in the repository in order to avoid creating a thermal perturbation.

Two of the listed fuels, U-Th metal with a NaK bond and MSRE fuel, contain reactive materials that must be destroyed in order to comply with the requirements of 10 CFR 60. In the case of the NaK-bonded fuels, this will require destruction of the cladding followed by reaction or removal of the liquid metal. The MSRE fuel must be treated in order to sequester the fluoride ion. One possible treatment for this fuel involves mixing the fuel salt with other ceramic-forming materials and then reacting and densifying the mixture in a hot isostatic press.

TABLE I

Miscellaneous Spent Fuel Characteristics

Fuel source	Fuel composition	Cladding material	Heavy-metal mass (Mg) On hand	Heavy-metal mass (Mg) Annual production	Burnup (GWd/Mg)	Displacement volume,[a] (m^3)	Heat generation rate[b] W/Mg	Heat generation rate[b] W/m^3	Heat generation rate[b] Total W
Peach Bottom 1/1	(U-Th)C	PyC[c]	1.4		37	19	1500	110	2,100
Peach Bottom 1/2	(U-Th)C	PyC	1.4		73	19	3000	220	4,200
Fort St. Vrain	(U-Th)C	PyC, SiC	8	1.5	16-50[a]	256	1700[b]	53	13,600[b]
Dresden	UO_2-ThO_2	SS	2.5		4-10	0.8	300-700	1560	1,250
Elk River	UO_2-ThO_2	SS	5		<50, 20 avg.	1.5	700	2300	3,500
LWBR	ThO_2-UO_2	Zr	20		<5 est.	6	<175	<580	<3,500
SRE	U-Th metal	SS, NaK bond	2.1		10	0.6	350	1200	700
EBWR	UO_2	Zr	1.6		1.6	0.5			
	UO_2-Zr	Zr	5		1.6	1.7	60	180	450
	UO_2-PuO_2		0.9		1.6	0.3			
HWCTR	UO_2	Zr	0.9		1 est.	0.3	35	100	30
Miscellaneous	UO_2	Zr	14		~10	5	~350	1000	~5,000
TRIGA	(U-Zr)H	Inconel, SS, or Al	0.2	0.02	20-40	0.75	1000	260	200
MSRE	(Li,Be,U)F	None	0.038	-	116	7	4000	21	150
Totals			63			310			35,000

[a]Applicable to material on hand.

[b]Calculated for decay time of 10 y.

[c]PyC = pyrolytic carbon.

Source: Ref. (13).

ACTIVATED METALS

Metals used in fuel assemblies and in other high-neutron-flux zones in a nuclear reactor will become radioactive, probably to such an extent that their disposal in a geologic repository will be required. This section discusses the available information concerning activated-metal characteristics and describes the packaging and containment systems that potentially might be employed for this class of radioactive waste.

The bulk of the activated metals will derive from spent fuel hardware, including BWR fuel channels and cruciforms. Assuming fuel consolidation, the estimated volumes of full density activated metals per MgU from the four major sources are as follows: PWR and BWR disassembly hardware ~0.006 m^3; BWR fuel channels ~0.035 m^3; and BWR cruciforms ~0.01 m^3 (15).

Characteristics

The important characteristics of activated metals are their radioactivity levels expressed both in absolute terms and in comparison with Class C limits, heat generation per unit volume, and corrosion resistance in the repository environment. The magnitudes of the values for radioactivity and heat generation are indicated in Table II. To provide perspective, Table II also indicates the radioactivity and heat generation of 1000-y-decayed LWR spent fuel. Substantial uncertainty exists concerning the values for characteristics of spent fuel disassembly hardware due to a lack of reliable data on minor constituents of the metals, the neutron flux, and the effective cross sections in areas outside the fueled region.

Four levels of metal activation are related to the metal source. The four generic sources are: (1) early PWR, (2) modern PWR and BWR fuel disassembly hardware, (3) BWR fuel channels, and (4) BWR cruciforms. On the basis of current information, all metals used in a fuel assembly in any zone, with the exception of the stainless steel present in the top end fitting, will be activated beyond the Class C limits of 10 CFR 61. If this is the case, there seems to be no incentive to segregate the more active components from the less active ones. The radioactivities cited in Table II are based on homogenization of all fuel assembly hardware.

The radioactivity source that dominates the contributions to multiples of Class C limit is Nb-94, which has a 20,000-y half-life. The parent of Nb-94 is natural niobium, which is present as an impurity in Zircaloy and stainless steel (as used in fuel assembly end pieces) and as a major constituent in the nickel-based alloys used in fuel assembly springs, both within the core zone as grids and in end pieces as holddown springs.

After a decay time of 10 y the heat generation of SFD hardware is dominated by 5.3-y half-life Co-60, which will decay in an additional 25 y to produce less heat per unit volume than does 1000-y-decayed LWR spent fuel. The heat generation of the remaining radionuclides in SFD hardware is insignificant from the standpoint of repository performance.

TABLE II

Radioactivity and Heat Generation of Activated Metals and LWR Spent Fuel

	Metal source				
	Early PWR hardware	Modern PWR or BWR hardware	BWR channels	BWR cruciforms	Spent fuel
Multiple of Class C LLW limit[a]	600	100	7	4	80,000
Radioactivity at 1000-y decay,[b]					
Ci/m^3					
C-14	40	20	9	37	
Ni-59	350	60	0.06	69	
Ni-63	26	5	0.005	6	
Zr-93	1	2	4	-	
Nb-94	120	20	1.4	0.4	
Total	540	110	18	110	5220[c]
Ci/Mg U	3	0.7	0.7	1.5	1740
Thermal output,[b] W/m^3					
10-y decay	(2800/5/1)2800[d]	(600/1/0.2)600[d]	35	563	3420[c]
35-y decay	110	20	0.9	22	
63-y decay	7	2	0.07	1	
1000-y decay	1	0.2	0.01	0.02	165[c]
Volume, m^3/MgU	0.0066	0.0066	0.04	0.013	0.33

[a]Assuming 10-y decay and that all SFD parts are blended. See 10 CFR 61 for definition of Class C.

[b]Full-density metal.

[c]3 MgU/m^3; burnup of 33 GWd/Mg U; data on radioactivity from ref. 14.

[d]Number sequence can be described as follows: (largely Co-60/Ni-63/largely Nb-94) total.

Sources: Refs. (13 and 15).

The metals used in fuel assembly hardware are exceptionally resistant to corrosion by the high-temperature water used in PWRs and BWRs. The temperature of the SFD hardware metals in a repository will be near ambient if the short-lived activity is allowed to decay prior to emplacement. The rate of corrosion will obviously be related to the fluid medium contacting the metal, which will vary with the nature of the repository in which the metal is emplaced. Early results on the corrosion of stainless steel indicate a corrosion rate in well water from Yucca Mountain of <0.2 μm/y (16). Inconel alloys generally yield corrosion rates in water comparable to those of stainless steel. Corrosion rates for stainless steel in polluted river waters have been observed to be less than 2.5 μm (17).

Applicability of 10 CFR 60 to Activated Metals

The NRC regulations applicable to HLW, 10 CFR 60, were obviously formulated with the intent of protecting the environment from fission product-containing and actinide-bearing wastes. The activated metals contain relatively small amounts of beta-gamma emitters and no alpha emitters except those that might be present due to trace quantities of uranium in the metals and to possible pickup of transuranics or fuel particles in the course of reactor operation and/or fuel disassembly.

As discussed earlier, the period of high heat generation in activated metals is due to the presence of Co-60, which has a 5.3-y half-life. This is in contrast to vitrified HLW and LWR spent fuel, whose heat generation after a 5 to 10-y decay period is largely due to the radionuclides with ~30-y half-lives. Because of the shorter half-life of Co-60 and the very stable nature of activated metals, it seems reasonable to allow the high-heat-generation source to decay prior to repository emplacement. In the absence of a high-heat-generation period, the requirement for complete containment appears to be invalid (assuming that nonheat producers are isolated in the repository from heat producers).

The annual fractional release rate for spent fuels and vitrified HLW is based on the presence of relatively large quantities of actinides and the associated long-lived fission products. Because the radioactivity of activated metals is due almost entirely to a few beta-gamma emitters, the appropriateness of employing the same release rate limit for activated metals as for conventional HLW appears to be questionable. Further evaluation seems to be in order.

The decay time at which the radioactivity of activated metals is calculated in order to determine the maximum permissible release rate apparently should be different from that for fission product-containing wastes. In view of the fact that heat production rates are quite low at 35 years, this decay time might be an appropriate base period. Radionuclides having half-lives less than 10-y might reasonably be exempted.

Modification and Packaging of Waste Forms

This subsection pertains to potential methods for satisfying the overall objective of the applicable radioactive waste disposal regulations. Previously, it was observed that some of the four possible generic process steps through which a waste may pass might be either deleted or combined with other steps if the original nature of the waste form and/or the repository environment is suitable.

Modification of the waste form will definitely be employed to achieve a reduction in the volume occupied by the as-produced activated metals. No chemical change in the waste form is feasible. The candidate methods for volume reduction are supercompaction, hot isostatic pressing, and melting and casting. These methods will achieve progressively higher densities. Their relative benefits and feasibilities are not clear.

As discussed previously, the absence of a thermal pulse or the repository conditions associated with significant areal heat loads in the repository seems to justify deletion of the requirement for substantially complete containment for any time period. Thus, the activated metals potentially can be placed in a repository in only a retrieval container.

The rate of corrosion and radionuclide transport of the activated metals will impact choices pertaining to modification of the waste form and design of the waste package. The allowable corrosion rate is apparently a function of both the metal surface area per unit mass and the allowable annual fractional release rate. If we assume that those metals which are the dominant sources of radioactivity release have an effective thickness of 1.0 mm and use an allowable annual release rate of 1 part in 10^5 of the quantity present at the time of emplacement, the allowable corrosion rate will be 5×10^{-6} mm/y, or 5×10^{-3} μm/y. This rate is based on unlimited removal of corrosion products from the scene as they are formed. For the more active hardware coming from early PWR fuel assemblies, the bulk of the radioactivity will apparently derive from grid spacers made of ~0.7 mm thick Inconel.

A corrosion rate of the order of 5×10^{-3} μm/y is probably unattain-able (see above). Two options appear to be possible for overcoming this problem. The first is to process the metals into much more massive pieces. Increasing the effective thickness of the waste form to 100 mm would alter the required corrosion rate to levels that probably would be attainable for at least some repository environments without providing special barriers for liquids. The second potential option is to employ a migration barrier around the waste form to reduce the effective corrosion rate by a factor of 100 to 1000.

TRANSURANIC WASTES (TRUW)

The anticipated major sources of TRUW from commercial power production operations are the fuel handling and consolidation facilities used in receiving spent fuel from the utilities and in preparing it for interim storage and/or repository emplacement. Very small amounts are normally generated by the nuclear reactors. A substantial amount will result from decommissioning of the West Valley Demonstration Plant. Because the TRUW resulting from the operations carried out in fuel handling and consolidation facilities will be a continuing significant source, it will receive exclusive consideration here.

In all cases, TRUW contains relatively small amounts of actinides mixed with a much larger mass of nonradioactive material. TRUW generated by an MRS or repository fuel handling facilities will consist of HEPA filters, and probably some prefilters, that have trapped a small amount of fuel particles, and a larger amount of nonradioactive dust. These fuel particles, which will be released from cladding failures, will contain not only transuranics but also the spectrum of fission products.

It is not known what fraction of the fuel entering the facility will reach the filters. We are inclined to estimate that 1 g of fuel per Mg of fuel handled is a conservative value. One estimate of waste volume (13) arrived at a final TRUW volume of ~0.02 m^3 per Mg of HM handled, which would be equivalent to 50 g of fuel per m^3. The heat release from this quantity of fuel at 10-y decay is ~0.05 W.

The NRC criteria with respect to complete confinement during the period of high heat generation appear to be inapplicable because of the absence of a period of high heat generation, provided the TRUW is stored substantially apart from those wastes which generate large amounts of heat. The long-term annual release criteria of 1 part per 10^5 appears to be automatically satisfied provided the following conditions exist: (1) the fraction of the fuel reporting to TRUW is not substantially greater than 1×10^{-5}, (2) the TRUW is considered as a displaced portion of the fuel assembly and (3) the fuel assembly containment is substantially complete for 300 to 1000 y. If long-term rate of release is a concern, the only viable option appears to be the use of a migration barrier around the waste form.

Because TRUW is expected to be finely divided, treatment (waste form modification) may be required to consolidate the particulates.

OXIDE CONTAINING U-233

About 1 Mg of uranium is presently being stored (13). Its approximate isotopic distribution, in percent, is as follows:

U-232	0.014
U-233	9.67
U-234	1.40
U-235	76.50
U-236	5.63
U-238	6.84

The total radioactivity of this material has been calculated and found to be comparable to the alpha activity of standard-burnup PWR fuel at 1000-y decay. Hence, on a mass basis, this source of actinide activity is comparable to LWR spent fuel.

The maximum heat generation was calculated to be 189 W/Mg U, occurring at ~50,000 y decay. This is to be compared with 55 W/Mg U for 33 GWd/Mg PWR fuel at 1000 y. The physical form of this uranium oxide is a solid cake deposited in a closed stainless steel tube (13).

There seems to be no reason to modify this waste form prior to its emplacement in a repository. Because of the absence of a period of high heat generation, there is apparently no need to provide substantially complete containment for any period, although in some repository settings the existing container will provide that function for a considerable length of time.

If there should be a demonstrable need to retard the rate of release of radionuclides from this oxide, the obvious approach would be to use a diffusion barrier.

SUMMARY

The EPA standards set forth limitations regarding releases to the accessible environment adjacent to a geologic repository. The NRC criteria pertaining to waste form and engineered barrier performance place certain restrictions on the physical and chemical nature of the waste form and require substantially complete confinement of radioactivity until the high-heat-production period is past. After this period, the annual release of radionuclides from the waste package is normally limited to 1 part in 100,000 of the amounts calculated to be present at 1000-y decay. The regulation permits deviation from these criteria in exceptional circumstances.

An exceptional circumstance which might justify deletion of the 1000-y container for a high-level waste is the absence of a significant perturbation in temperature around (and possibly but apparently not necessarily in) the stored waste. The lack of significant heat release will eliminate the hydrologic driving force for dispersal of radionuclides. Exceptional circumstances which potentially could justify a less stringent long-term release criterion are: (1) small quantity of radioactivity, (2) the nature of the radioactive species, and (3) the nature of the geology in which the waste is to be emplaced.

Because the MW after a suitable decay period have low heat release rates per unit volume, they apparently could be so emplaced in a repository that there would be no compelling need, according to the reasoning presented in 10 CFR 60, for a 1000-y container. Regarding attainment of the specified long-term release rate criterion, neither the solubility limits for the various waste forms nor the conductance of potential migration barriers are currently adequately characterized. The relatively small total heat generation rate for the MW in combination with the usual low volumetric heat generation rate apparently will allow application of migration barriers in a low temperature environment where barrier performance would be expected to be unchanged with time. Further studies will be required to determine the optimal distribution of protection to be afforded by the various elements of the engineered barrier system.

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