RADIOISOTOPE INVENTORY FOR TSPA-SR

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INTRODUCTION

The total system performance assessment for site recommendation[1] (TSPA-SR), on Yucca Mountain, as a site (if suitable) for disposal of radioactive waste, consists of several models. The Waste Form Degradation Model (i.e., source term) of the TSPA-SR, in turn, consists of several components.[2,3] The Inventory Component, discussed here, defines the inventory of 26 radioisotopes for three representative waste categories[4]: (1) commercial spent nuclear fuel (CSNF), (2) U.S. Department of Energy (DOE) spent nuclear fuel (DSNF), and (3) high-level waste (HLW). These three categories are contained and disposed of in two types of waste packages (WPs)—CSNF WPs and co-disposal WPs, with the latter containing both DSNF and HLW.[4] Three topics are summarized in this paper: First, the transport of radioisotopes evaluated in the past; second, the development of the inventory for the two WP types and third, the selection of the most important radioisotopes to track in TSPA-SR.

PREVIOUS INVENTORY ESTIMATES AND SCREENING OF RADIOISOTOPES

Assessments of Yucca Mountain in the early 1990s based the inventory for HLW and CSNF[6] on the DOE Characteristic Data Base (CDB) of 1987[5], which was developed about the time of the 1988 Site Characterization Plan.[7] In 1991, 39 radioisotopes were identified as potentially important based only on CSNF for TSPA-91.[8,9] All 39 radioisotopes were included in the human-intrusion (or direct release) and volcanism scenarios; however, only nine radioisotopes thought to be the major contributors to releases were included in the groundwater (GW) release scenario, since more complicated models were used in this scenario.

In the TSPA conducted in 1993 by Sandia National Laboratories (TSPA-93-SNL),[10] 43 radioisotopes were identified as potentially important based on whether the inventory of both CSNF and HLW—normalized to the release limits established by the U.S. Environmental Protection Agency (EPA) in 40 CFR 191[11]—contributed at least some fraction of the total cumulative release over the period from 10^3 to 10^6 years. All 43 radioisotopes were included in the human-intrusion and volcanism scenarios; however, only eight radioisotopes were included in the groundwater scenario. In the TSPA conducted in 1993 by Intera (now Duke Engineering Services) (TSPA-93-Duke),[12] the CDB of 1992[13] was used for CSNF inventory. In TSPA-93-Duke,[12] both 40 CFR 191 (based on population dose) and individual dose were used to determine the potential contribution of each radioisotope over a period up to 10^6 yr. For these calculations, a simple spreadsheet was used to evaluate radioisotope escape, dilution, and ingestion. Any radioisotope that contributed more than 0.005% to the total dose was included. A total of 37 radioisotopes were selected and used in all three basic scenarios (since simple models were used in all scenarios). The total contribution to dose from radioisotopes that were excluded was 0.01%.

TSPA-95[14] used the CDB of 1993[15] for CSNF where the weighted average burnup was slightly lower than the CDB of 1992 (36,666 versus 39,075 MWd/MTHM [mega watts day per metric tonnes of heavy metal][14]) but still used the CDB of 1987 for HLW. For selecting radioisotopes, a procedure similar to that used for TSPA-93-Duke was used; 39 radioisotopes were selected and used in all three basic scenarios. Also in 1995, the U.S. Nuclear Regulatory Commission (NRC) performed an Iterative Performance Assessment (IPA).[16] A radioisotope was retained in the IPA if it contributed more than 1% of the EPA release limit for that radioisotope from 40 CFR 191.[11] The screening analysis also checked the maximum dose to a farm family to determine whether any of the radioisotopes that might have been screened out on the basis of cumulative release should have been retained on

the basis of individual dose; 19 radioisotopes were selected (Table 1).

For the TSPA conducted for the viability assessment, (TSPA-VA)[17,18] a radioisotope was included if it had: (1) a high solubility, (2) a low sorption affinity, (3) a significant inventory, (4) a high dose conversion factor, (5) a long half-life, and (6) potential for colloidal transport. The same 39 radioisotopes used in TSPA-95 were available in the source term, but only nine radioisotopes were tracked in the groundwater scenario because more complicated transport models were once again used in the unsaturated and saturated zones (Table 1). In addition, two sensitivity studies examined the influence of (a) Pu disposition wastes[19] and (b) 11 DSNF types.[17,20]

INVENTORY FOR TSPA-SR

TSPA calculations for the site recommendation and proposed new regulations for Yucca Mountain by NRC and EPA[24] have prompted a new evaluation of important radioisotopes to monitor as discussed below.

Basis for CSNF Inventory

As currently projected, about 230,000 CSNF assemblies will be disposed in the potential repository at Yucca Mountain.[4] Average isotopic compositions were developed using the most up-to-date historical data from utility companies on assembly discharges from their reactors through December 1995. The utilities have also provided a forecast for the assembly discharges over the next five reloading cycles, which occur about every 1.5 years (~June 2002). This CSNF data includes the trend toward burning the fuel for longer periods in the reactor. Based on the information, average and bounding radioisotope activities per assembly were estimated, and the WP that could accommodate the assembly based on a criticality criterion was determined. The result was a grouping of the CSNF assemblies into five WP groups. The inventory for TSPA-SR, was then calculated based on the estimated number of packages for each group. The two largest groups out of the five were 4500 packages containing 21 assemblies of pressurized water reactor (PWR) fuel and 3000 packages containing 44 assemblies of boiling water reactor (BWR) fuel (Figure 1). For selecting radioisotopes to track in the TSPA-SR, the activity per assembly of four assemblies (an average and bounding PWR and an average an bound BWR) was used in the screening procedure described in the next major section.

Basis for DSNF Inventory

The TSPA-SR analysis used an updated inventory for DSNF.[4,21,22] The DSNF waste form category encompasses more than 250 distinct types of spent fuel, with radioisotope inventories that vary widely, depending on the reactor history of the fuel.[22] The DSNF waste form will be packaged in three types of canisters (consisting of one multi-canister overpack for N-Reactor SNF and two canisters of different lengths[a]) before they are shipped to the potential repository for disposal. For evaluating the activity of DSNF in TSPA-SR, an average radioisotope activity for the DSNF waste form was calculated by using the estimated activity for each of the three canister types.[b] For radioisotope screening discussed below, the total inventory of all ~250 waste types was used to determine an average and the inventory for **virgin** SNF was used to represent a bound (Figure 2).

Basis for HLW Inventory

HLW in borosilicate glass is currently produced and stored at the Savannah River Site (SRS), South Carolina. Production of HLW glass is also anticipated to start at the Hanford Site, Washington, and the Idaho National Engineering and Environmental Laboratory (INEEL), Idaho. Finally, a small amount of HLW glass was produced at West Valley (WV), New York, but the exact amount to be disposed has not been finalized. These generator sites provided radioisotope inventories for the HLW representative of their vitrification process. This information, summarized in the draft environmental impact statement (DEIS) on Yucca Mountain,[23] was used to calculate an average radioisotope inventory for the short and long HLW canisters. The inventory of excess plutonium waste was also included in the HLW (Figure 2). For radioisotope screening discussed below, the total inventory of all HLW canisters was used to determine an average and the inventory for SRS was used to represent a bound.

RADIOISOTOPE SCREENING FOR TSPA-SR

The radioisotopes for the eight waste categories (average and bounding, PWR, BWR, DSNF, and HLW) was screened to identify radioisotopes that might make significant contributions to the expected annual dose without depending on past TSPA results. Similarly to TSPA-95, the relative importance was determined using a simple spreadsheet. The screening considered the following three factors[4]: (1) relative inventory in any one period (radioisotope initial inventory and longevity or production); (2) dose conversion factor (contribution to both ingestion or inhalation dose); and (3) transport

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[a] The canisters may also be of different diameters, but this variation was not considered initially.

[b] The Naval spent nuclear fuel (SNF) will be placed in two types of canisters (consisting of two lengths) and placed in a waste package without any HLW. Analysis of Naval SNF is discussed elsewhere.[3]

sorption affinity[c]. In considering the sorption affinity of a radioisotope, factor 3 the radioisotopes were divided up into three groups: (a) high sorption (thus release via colloidal transport necessitated, (b) moderate sorption (thus release via fraction flow dominates), and (c) low sorption (thus release not constrained except by solubility). The importance of radioisotopes was then evaluated separately. Those radioisotopes that made up 95% of the total dose when considering factors 1&2 for the vulcanism scenario, and the union of factors, 1,2&3a, 1,2&3b, and 1,2&3c for the human-intrusion and GW scenario for two periods (10^4 and 10^6 yr) were included. A total of 23 radioisotopes was selected (Table 1).

This initial list of radioisotopes was then augmented to account for in-growth of the actinide decay chains. For example, for the period of 10^6 yr, ^{235}U was added to the list because ^{227}Ac, which is potentially important to dose, is one of its daughters. In addition, ^{228}Ra and its precursor, ^{232}Th, were added because of their relevance to the groundwater protection requirement in the EPA proposed standard.[24] These adjustments expanded the list to 26 radioisotopes for the TSPA-SR (Table 1).

IMPLEMENTATION IN TSPA-SR

The computer implementation of the inventory abstraction is a simple table look-up of the quantity of radioisotopes at the time of waste emplacement for the CSNF and co-disposal WPs. An overall average radioisotope inventory for all the CSNF WPs was computed by weighting by the estimated number of packages in each of the five WP groups to reach 63,000 MTHM (90% of 70,000 MTHM repository)[25,26] (Figure 1). The average inventory for a co-disposal package was also developed from average inventory for the six co-disposal configurations to reach 2333 MTHM of DSNF (3.3%) and 4667 MTHM HLW (6.7%) (Figure 2). As currently modeled, there are 7860 CSNF WPs and 3910 co-disposal WPs[4]. These activities are adjusted for decay, in-growth, and release from the waste form during the TSPA-SR simulations.

While most radioisotopes in the CSNF waste are bound in the UO_2 matrix, some fission product gases such as ^{137}Cs and ^{129}I are known to migrate to the gap between the matrix and cladding while in the reactor. Furthermore, many radioisotopes can migrate from the matrix to the cooler grain boundary. This migration process is important because these radioisotopes are released much faster than those bound in the fuel matrix. To account for distinct release rates, the inventory for the CSNF waste form is divided into two parts: (1) matrix and (2) fast release. The fast release inventory is the sum of the gap inventory and grain boundary inventory. The grain boundary inventory is assumed to be the total inventory of each radioisotope times a fraction sampled from a uniform distribution between 0 and 0.004. The distribution is based on the fractions of radioisotopes observed to be released in short-term tests (less than 200 days) and extrapolated to 5 years.[3,27,28] The gap inventory is an additional 0.042 and $1/3 \times 0.042$ of the inventory of ^{137}Cs and ^{129}I, respectively.

UNCERTAINTY AND VARIABILITY OF INVENTORY

The radioisotope inventory used in TSPA-SR is deterministic and does not include statistical distributions to express data uncertainty or variability. The principal source of uncertainty and variability in the current inventory analysis is in the projections of final waste form inventories. The actual waste streams may differ with respect to fuel burn-up, fuel age, fuel enrichment, and reactor efficiency. However, of the 230,000 CSNF assemblies modeled in the TSPA-SR, the activity of important radioisotopes vary only by an order of magnitude. More importantly, the variation of activity in the CSNF packages is much less than the variation in other components (e.g., CSNF cladding unzipping rates have a three-orders-of-magnitude variation[1,27,28]). The range in the co-disposal packages varies more (the range in the HLW canisters varies by two orders of magnitude and in the DSNF canisters by five orders of magnitude), but the total inventory is so much smaller in the co-disposal packages. As shown in Figure 3 and the total number of WPs of each type, the total inventory of ^{237}Np and ^{99}Tc in co-disposal packages is more than an order of magnitude less than in CSNF packages. Hence, the CSNF packages have much greater influence on the dose, provided the release rates are similar—which they are.[3,7] Furthermore, DSNF contributes only 2% of the ^{237}Np and ^{99}Tc inventory in the co-disposal packages (Figure 3). Hence, variability of the inventory in the co-disposal packages and especially DSNF can be much greater and still not influence the total dose.

The current inventory analysis for TSPA-SR is more detailed and flexible than those done for previous TSPAs. With regard to the selection of important radioisotopes, the radioisotope screening procedure is considered sufficiently conservative in that it identifies a larger set of radioisotopes than would actually be needed to appropriately determine the expected annual dose.

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[c] A fourth factor, elemental solubility (i.e., low solubility for Am, Cm, **Zr**, Th, Nb, **Pa** and Sn) was also considered but the union of 1,2,&4 with 1,2,&3 did not add any additional radioisotopes.

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Table 1. Radioisotopes transported in TSPA-SR scenarios and past analyses

Isotope	TSPA-SR						TSPA-VA GW	NRC 99	TSPA 93&95
	GW		Volcanic		Intrusion				
	10^4	10^6	10^4	10^6	10^4	10^6			
Total	16	21	12	16	18	23	9	19	38
^{14}C	•	•			•	•	•	•	•
^{99}Tc	•	•			•	•	•	•	•
^{129}I	•	•			•	•	•	•	•
^{227}Ac	•	•	•	•	•	•			•
^{229}Th	•	•	•	•	•	•			•
^{230}Th		•		•		•		•	•
^{232}Th									•
^{232}U	•	•	•	•	•	•			
^{233}U	•	•	•	•	•	•			•
^{234}U	•	•			•	•	•	•	•
^{235}U									•
^{236}U	•	•			•	•			•
^{238}U	•	•			•	•		•	•
^{237}Np	•	•			•	•	•	•	•
^{238}Pu	•	•	•	•	•	•			•
^{239}Pu	•	•	•	•	•	•	•	•	•
^{240}Pu	•	•	•	•	•	•		•	•
^{242}Pu		•		•		•	•		•
^{241}Am	•	•	•	•	•	•		•	•
^{243}Am	•	•	•	•	•	•		•	•
^{90}Sr			•	•	•	•			
^{137}Cs			•	•	•	•		•	
^{210}Pb		•		•		•		•	•
^{231}Pa		•	•	•		•	•		•
^{226}Ra		•		•		•		•	•
^{228}Ra									•
^{36}Cl									•
^{59}Ni									•
^{63}Ni									
^{79}Se							•	•	•
^{93m}Nb									•
^{94}Nb								•	•
^{99}Zr									•
^{107}Pd									•
^{126}Sn									•
^{135}Cs								•	•
^{151}Sm									•
^{241}Pu									•
^{242m}Am									•
^{244}Cm									•
^{245}Cm								•	•
^{246}Cm								•	•

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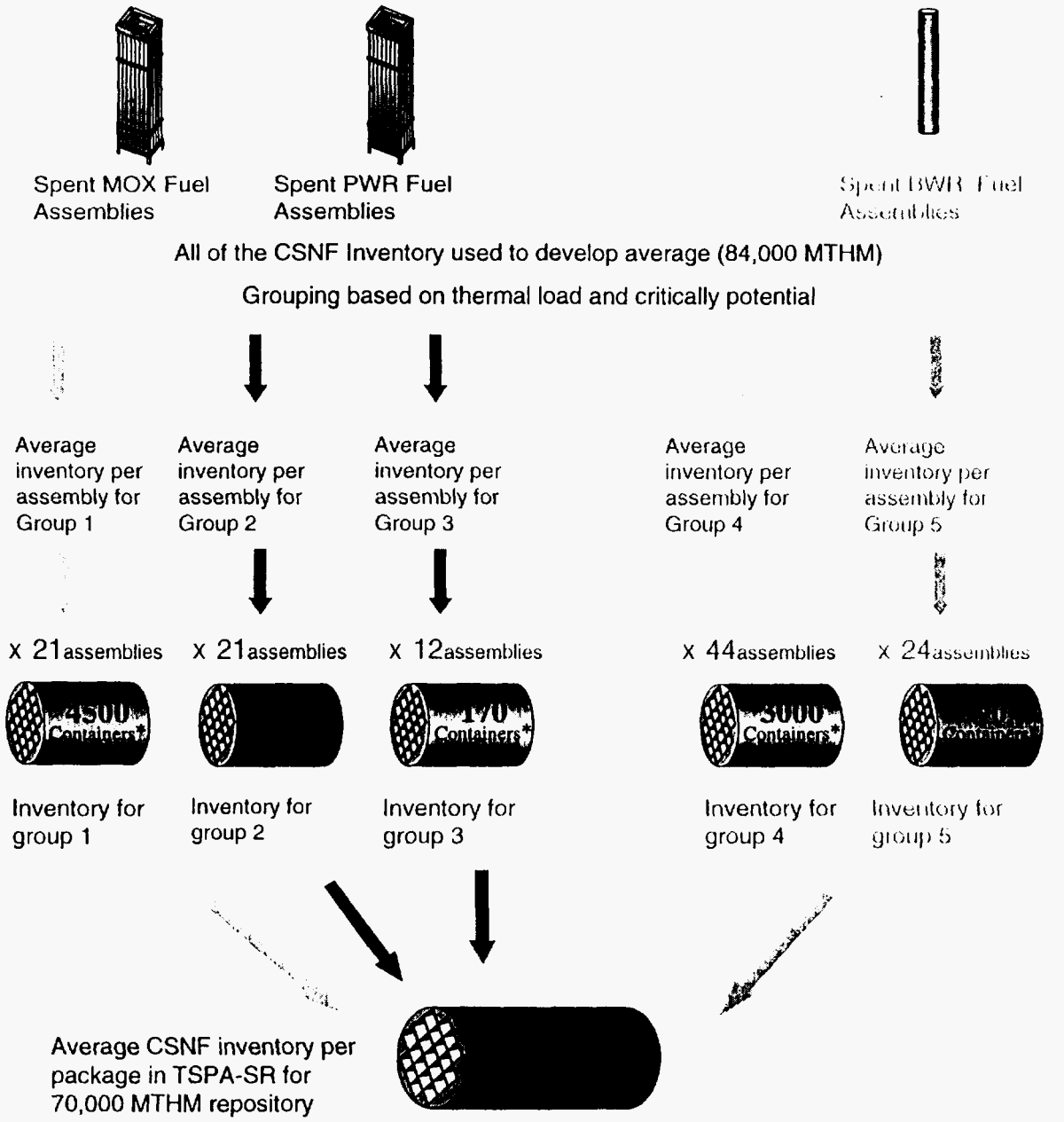

Figure 1. Radioisotope activity in CSNF packages based on average inventory developed for five groups

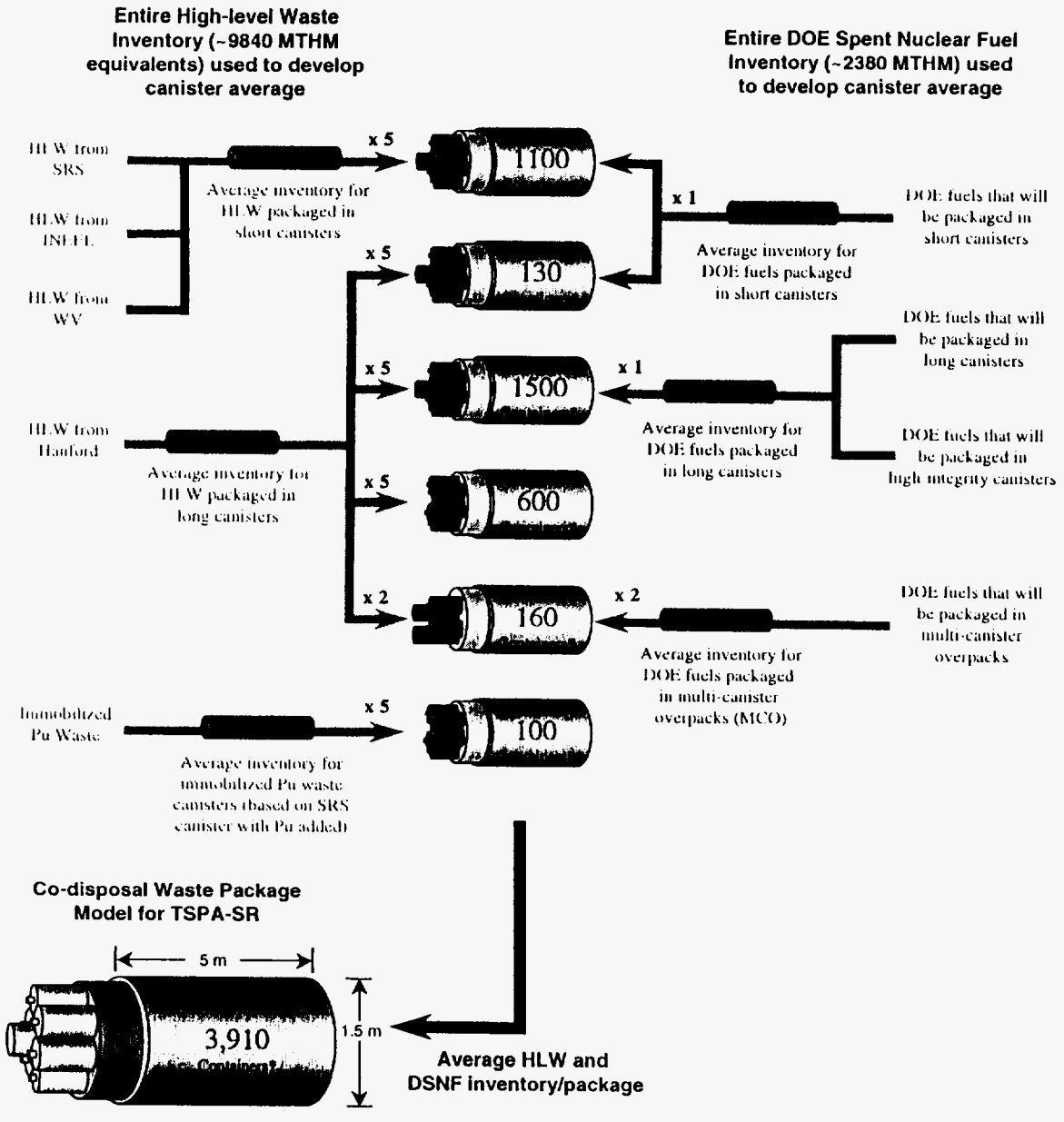

Figure 2. Radioisotope activity in co-disposal packages based on average inventory in several groups

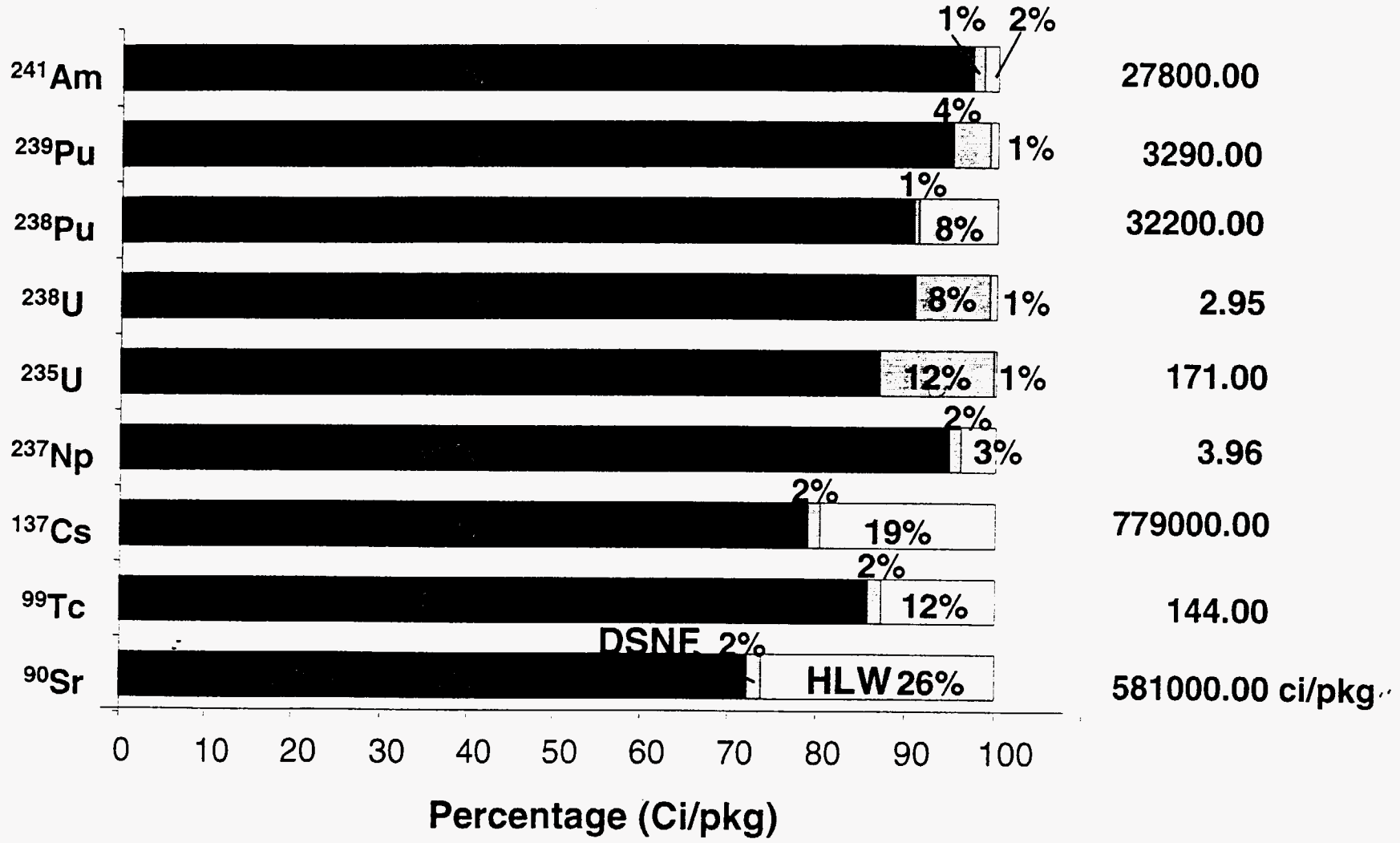

Figure 3. Highest percentage of radioisotopes reside in CSNF packages