EFFECTS OF ACTINIDE COMPOSITIONAL VARIABILITY IN THE U.S. SPENT FUEL INVENTORY ON PARTITIONING-TRANSMUTATION SYSTEMS

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The partitioning and transmutation concept (P-T) has as a mission the reduction, by many orders of magnitude, of certain undesirable nuclides in the waste streams. Given that only a very small fiction of spent fuel can be "rejected" by a P-T enterprise, a P-T system must therefore be capable of accommodating a wide range of spent fuel characteristics. Variability of nuclide composition (i.e. the feed material for transmutation devices) may be important because virtually all transmutation systems propose to configure TRU nuclides recovered from discharged LWR fuel in critical or near-critical cores. To date, all transmutation system core analyses assume nonvariable nuclide concentrations for startup and recycle cores. Using the Department of Energy's (DOES) Characteristic Data Base (CDB) and the ORIGEN2 computer code, the current and projected spent fuel discharges until the year 2016 have been categorized according to combinations of fuel burnup, initial enrichment, fuel age (cooling time) and reactor type (boiling-water or pressurized-water reactor). In addition to quantifying the variability of nuclide composition in current and projected LWR fuel discharge, the variability of the infinite multiplication factor (k_∞) is calculated for both fast (ALMR) and thermal (accelerator-based) transmutersystems. It is shown that actinide compositional variations are potentially significant and warrant further investigation.

'University of California, Berkeley under appointment to the Civilian Radioactive Waste Management Fellowship program administered by Oak Ridge Associated Universities for the U. S. Department of Energy.

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ABSTRACT

Partitioning and transmutation (P-T) is an advanced waste management concept by which certain undesirable nuclides in spent fuel are first isolated (partitioned) and later destroyed (transmuted) in a nuclear reactor or other transmutation device. here are wide variabilities in the nuclide composition of spent fuel. This implies that there will also be wide variabilities in the transmutation device feed. As a waste management system, P-T must be able to accept (all) spent fuel. Variability of nuclide composition (i.e., the feed material for transmutation devices) may be important because virtually all transmutation systems propose to configure transuranic (TRU) nuclides recovered from discharged light-water reactor (LWR) spent fuel in critical or near-critical cores.

To date, all transmutation system core analyses assume invariant nuclide concentrations for startup and recycle cores. Using the U.S. Department of Energy's (DOE'S) Characteristics Data Base (CDB) and the ORIGEN2 computer code, the current and projected spent fuel discharges until the year 2016 have been categorized according to combinations of fuel burnup, initial enrichment, fuel age (cooling time) and reactor type (boiling-water. or pressurized-water reactors). In addition to quantifying the variability of nuclide composition in current and projected LWR fuel discharges, the variability of the infinite multiplication factor (k_∞) is calculated for both fast (ALMR) and thermal (accelerator-based) transmute systems. It is shown that actinide compositional variabilities are potentially significant and warrant further investigation.

INTRODUCTION

Partitioning and transmutation (P-T) is a concept that greatly reduces the amounts of long-lived radionuclides in wastes going to a repository. Radionuclides that would be destroyed by P-T are generally considered to be the transuranic (TRU) actinides (i.e., Np, Pu, Am, and Cm) and certain long-lived fission products, such as ^{129}I and ^{99}Tc. Implementation of the P-T concept would involve intensified processing (partitioning) to remove long-lived radionuclides from the waste stream and subsequent use of a transmutation device to convert the actinides to fission products and the fission products to short-lived radionuclides. Fast-spectrum liquid-metal-cooled reactors (LMRs) are most often suggested for

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transmuting actinides, and thermal-spectrum devices are usually suggested for fission product transmutation.

Several different concepts for P-T of high-level radioactive waste (HLW) are currently being proposed or under investigation in the United States. Partitioning technologies for light-water reactor (LWR) spent fuel include both aqueous processes' and pyrochemical techniques.' Transmutation technologies include traditional reactor concepts, such as LWRS employing mixed-oxide (MOX) fuel;[3] fast reactors such as the Advanced Liquid Metal Reactor (ALMR);[4] subcritical devices driven by accelerator production of neutrons, such as the Phoenix Concept;[5] and the Accelerator Transmutation of Waste (ATW)[6] System. Most of the assessment and research performed to date by technology advocates has focused on the fundamental processes or the system design of the transmutation device or partitioning process rather than the integration of a P-T system into the existing commercial LWR fuel cycle in the United States.

Partitioning and transmutation were extensively evaluated over a decade ago by U.S. and foreign investigators. The conclusion was that the cost and short-term risk increases resulting from P-T substantially outweighed the long-term reduction in repository risk. A recent paper by Croff[7] re-examined the incentives for actinide P-T and identified a number of factors that have changed in the intervening years. Factors identified included (a) expectations concerning the ease with which a repository could be sited and licensed, (b) issuance by regulators of a repository licensing standard and supporting criteria, (c) the scenario to which P-T is compared, and (d) new technologies for fuel and waste processing. In a recent overview of P-T technologies,' it was noted that P-T has experienced a worldwide resurgence.

Activity at Oak Ridge National Laboratory has been directed at defining and describing the integration issues associated with a nuclear fuel cycle flow sheet employing P-T technology. Independent of the specifics of a technology, all P-T systems would have common interface points with the U.S. nuclear fuel cycle. A recent paper by Michaels[9] identified three primary interfaces between a generic P-T technology and the U.S. nuclear fuel cycle (1) the LWR spent fuel inventory, ('2) the reprocessed uranium (RU) stream, and (3) the HLW streams. Michaels provided some qualitative observations about the potential impact of nuclide variability within the U.S. spent fuel inventory on P-T systems. The objective of thii paper is to provide a preliminary assessment to quantify the effect of actinide compositional variability within the U.S. spent fuel inventory as it relates to the performance of representative P-T systems.

PROBLEM STATEMENT

Partitioning and transmutation (P-T) is an advanced waste management concept by which certain undesirable nuclides in spent fuel are first isolated (partitioned) and later destroyed (transmuted) in a nuclear reactor or other transmutation device. here are wide variabilities in the nuclide composition of spent fuel. This implies that there will also be wide variabilities in the transmutation device feed. As a waste management system, P-T must be able to accept (all) spent fuel. Variability of nuclide composition (i.e., the feed material for transmutation devices) may be important because virtually all transmutation systems propose to configure TRU nuclides recovered from discharged LWR spent fuel in critical or near-critical cores. To date, all transmutation system core analyses assume constant nuclide concentrations for startup and recycle cores, implicitly assuming that the as-loaded composition and reactivity of a fuel pin, assembly, and/or fuel batch can be tightly specified and well controlled.

However, the U.S. spent fuel inventory is neither homogeneous nor well-blended. Nuclide compositions in spent fuel can be expected to be a function of

- fuel bumup,
- initial emichment,
- reactor type (BWR or PWR), and
- age of the fuel (cooling time since discharge).

The variability in these parameters will, in turn, cause variability in the composition of the spent fuel. Thii variability in nuclide composition will need to be accommodated in the design (and licensing) of any transmutation system.

APPROACH

Each assembly within the LWR spent fuel inventory can be characterized in terms of its (a) fuel burnup, (b) initial emichment, (c) reactor type, and (d) cooling time since discharge. Using these characteristics, it is possible to calculate the radionuclide composition of each spent fuel assembly. Because thii would represent hundreds of thousands of separate calculations, fuel assemblies with similar characteristics have been grouped, reducing the number of calculations required to establish the overall compositional variability of the U.S. spent fuel inventory to about 300.

In thii investigation, we have assumed that large-scale deployment of P-T technology will occur in the year 2018 and will utilize the stockpile of commercial LWR spent fuel discharged from between the years 1968 and 2016 as feed material for the P-T system. The characteristics of the U.S. spent fuel inventory, as provided in the U.S. Department of Energy's (DOE's) Characteristics Data Base[10] (CDB) and its description of historical and projected U.S. spent fuel discharges, was utilized to determine the expected characteristics of the entire inventory of discharged spent fuel in the year 2018. P-T systems would be required to utilize the TRU actinides (Np, Pu, Am, Cm, etc.) recovered from LWR spent fuel as nuclear fuel within the transmutation device. Some transmutation devices may also utilize the reprocessed uranium (RU) as well.

The inventory of fuel assemblies projected to be in the U.S. spent fuel inventory in the year 2018 was sorted by its characteristics and divided into 1200 bins, with each bin representing a particular set of fuel burnup values, initial fuel enrichment, fuel age or cooling time, and reactor type [pressurized-water reactor (PWR) or boiling-water reactor (BWR)]. The CDB data are reported in terms of reactor type (BWR or PWR), discharge year, burnup bin (12 bins of 5000 MWd/MTIHM), average burnup, average enrichment, number of assemblies, and discharge mass. Historical data are those reported on the DOE's Energy Information Administration RW-859 data sheet." The projected data assume a scenario of "no new orders with extended burnup." For historical and projected spent fuel discharges through the year 2016, this scenario represents the discharge of nearly 250,000 spent fuel assemblies and 71,000 metric tons of spent fuel.

Using a personal computer (PC) spreadsheet program, the discharge data were grouped into burnup and enrichment bins. The maximum variation in initial fuel enrichment was 0.5% wide within each burnup bin. The total discharge weight, average burnup, and average enrichment was calculated for each bumup-enrichment bin combination. The total discharge weight (in units of metric tons) within each bin was determined by summing the spent fuel discharges between 1968 and 2016. The discharge year was used to redistribute the discharge weights according to 10 average cooling time bins. The result was 100 (50 PWR and 50 BWR) representative cases, with each case accounting for up to 10 different cooling times. These cases would provide 1000 spent fuel compositions, each unique in terms of the burnup, enrichment, type of reactor, and cooling time. When matched to the "actual" discharge data at

each combination of above parameters, the number of spent fuel compositions to be calculated can be reduced from 1000 to 300.

Using the ORIGEN2 computer code[12] and cross-section libraries[13] for standard- and extended-burnup PWRS and BWRs, the nuclide compositions for the 300 combination of burnup, initial enrichment, and cooling time were calculated. An example of a parameter of interest tos ystem designers is the relative quantity of minor actinides (defined as Np, Am, and Cm) to the total quantity of fissile plutonium nuclides (^{239}Pu and ^{241}Pu) in the LWR spent fuel. The quantity of minor actinides, which are net neutron absorbers, is significant for several reasons. It effects the neutron economy and the swing in reactivity during irradiation (the "burnup reactivity swing"). Figure 1 shows the variability of the mass ratio of the minor actinides to the fissile plutonium component in the LWR spent fuel inventory in the year 2018 (discharges through 2016). It should be noted that the mass ratio of the minoractinides is function of fuel burnup, and that increases in burnup correlate to increases in the amount of minor actinides in the fuel.

Reprocessing of LWR spent fuel produces three major material streams: (1) RU, (2) recovered TRUs, and (3) HLW. The TRUS provide the fissile material for the transmutation system. The disposition of the RU stream could involve long-term storage, disposal as a waste stream, or re-enrichment and recycle as LWR fuel. Complex institutional issues are involved in all these options. Some transmutation systems (particularly the ALMR) could use some of the RU as part of the reactor fuel. Figure 2 shows the variability in the ^{235}U assay within the U.S. spent fuel inventory. It should be noted that the majority of the ^{235}U assay variability in spent fuel falls within a fairly narrow range (between 0.7 and 0.9%). This is primarily due to the utility goal of fully recovering the economic value of each fuel assembly. The narrow range in the ^{235}U assay at discharge also means that the RU would exhibit fairly consistent neutronic characteristics if it is employed as part of the fuel within a transmutation system, such as the ALMR. However, nontrivial quantities of RU will be in the inventory at assays of between 1.0% and 2.0%. These higher assay RU streams originate principally from reactor campaigns that achieved fuel burnup levels that were lower than planned, such as in the case of defective fuel discharged early. Assemblies discharged early contribute to the overall variability in the RU stream and may be particularly important in transmutation systems that utilize relatively small batch sizes.

'he variability in LWR fuel TRU composition shown in Fig. 1 appears to be large. The question is: Will it be difficult for a P-T system to deal with this level of variability? The variability could pose a problem in design and licensing of the transmutation system. The infinite multiplication factor (or k_∞) may be used to quantify the relative impact that variations in the composition of TRU nuclides (i.e., approximately 40 isotopes of the elements Np, Pu, Am, Cm, Bk, Cf, Es) have on the as-loaded reactivity of the transmute system fuel. To quantify the impact, we have examined the value of k_∞ for each "batch" of TRUS placed in a reactor, where a batch is selected for its unique characteristics of burnup, enrichment, reactor type and cooling time. We assume that each batch does not alter the neutron spectrum within the core, when compared with a constant composition, homogeneous system. This assumption allows us to use the relative variability in the value of k_∞ to exhibit the impact "that the variability in the TRU composition would have on the fuel reactivity within a transmutation device. Ideally, k_∞ is defined as follows:

$$k_\infty = n'/n, \quad (1)$$

where n' is the number of neutrons in the present generation, and n is the number of neutrons in the previous generation. Therefore, the ratio n'/n is the ratio of neutrons produced to neutrons destroyed (or captured). In 0RIGEN2, this value is calculated on a unit basis by dividing number of neutrons

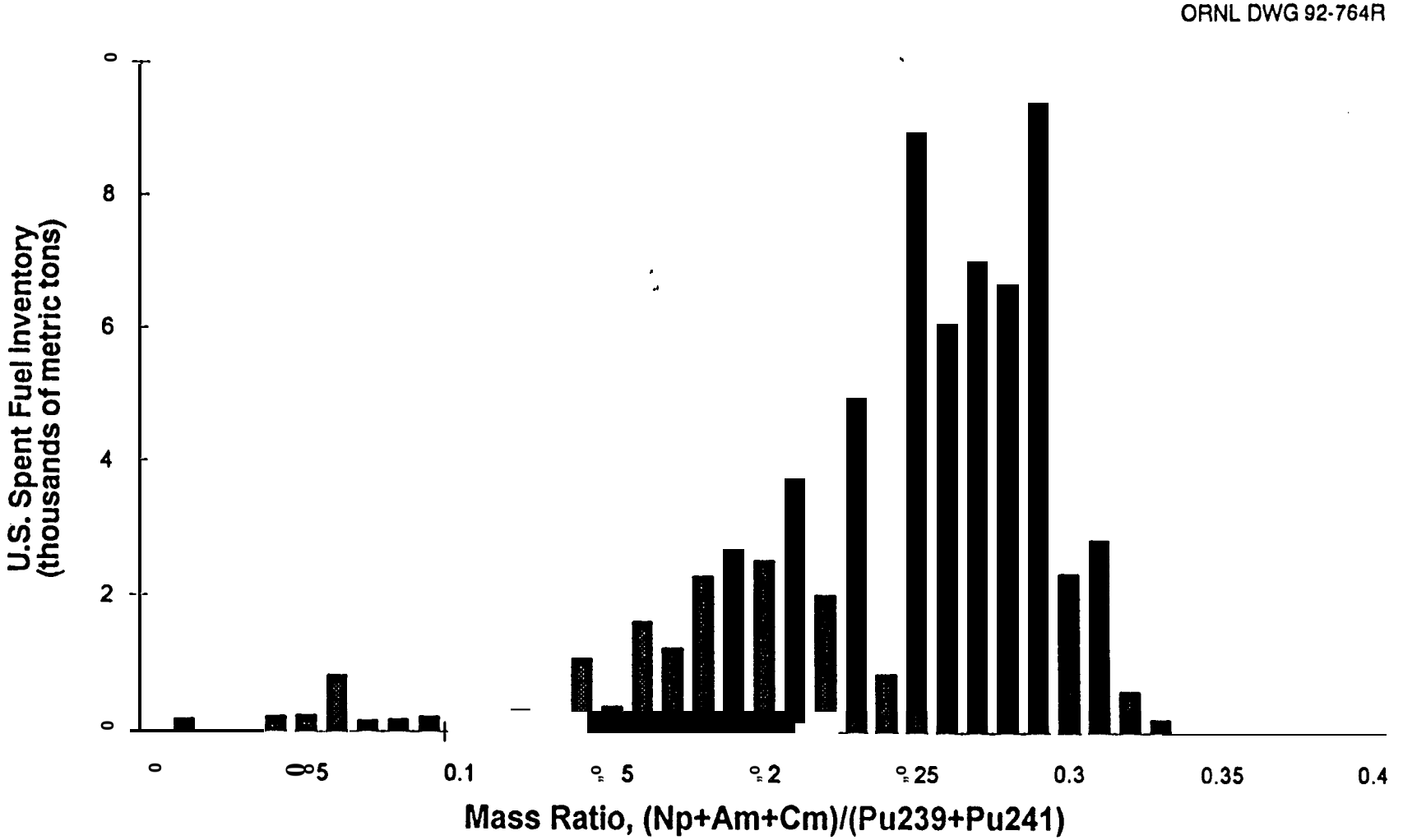

Fig. 1. Ratio of minor actinides to fissile Pu in all U.S. LWR fuel produced between 1968-2016 by 2018.

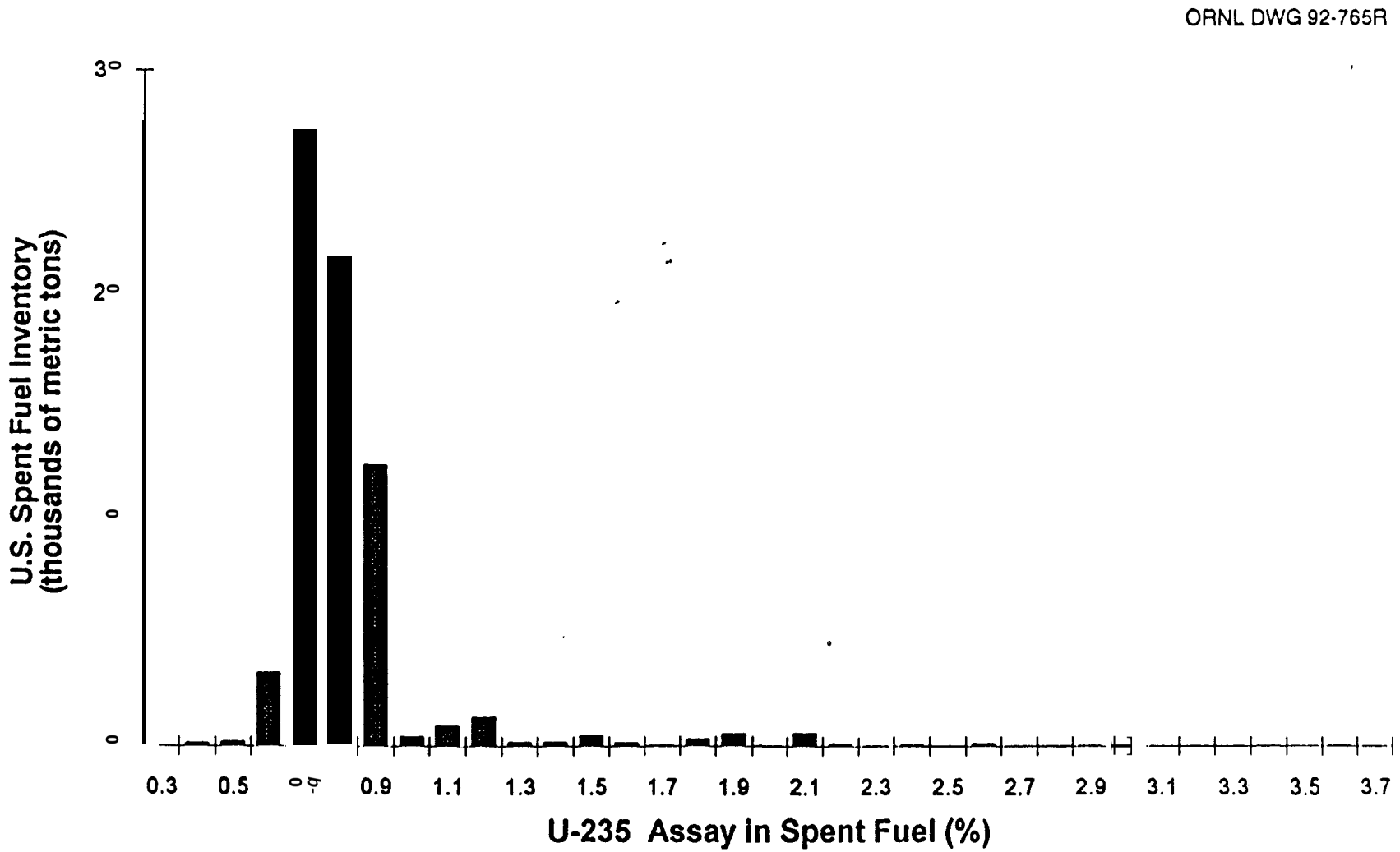

Fig. 2. Fraction of U-235 in total U for U.S. LWR spent fuel produced between 1968-2016 by 2018.

produced times the probability of neutron production by probability of neutron capture. The probabilities of production and capture are represented by the neutron cross section σ for the neutron-induced reactions of each TRU nuclide i. Rewriting, Eq. 1 may be represented as follows:

$$k_\infty = \Sigma (N_i * m_i) / \Sigma (\sigma_{T,i} * m_i), \quad (2)$$

where: N_i = neutron production cross section,

$= \sigma_{(n,f)} * \nu_f + 2.0 * \sigma_{(n,2n)} + 3.0 * \sigma_{(n,3n)}$;

m_i = mass (gram-atoms);

$\sigma_{T,i}$ = total capture cross section,

$= \sigma_{(n,\gamma)} + \sigma_{(n,2n)} + \sigma_{(n,3n)} + \sigma_{(n,f)} + \sigma_{(n,\gamma')} + \sigma_{(n,2n')}$; and

ν_f = neutron yield per neutron-induced fission.

In order to calculate the values of k_∞ for each combination of enrichment, burnup, and decay time would have required up to 300 additional ORIGEN2 calculations for each type of transmute system, with each calculation using the results of each individual LWR calculation as input. Of interest are systems based on fast neutrons, such as the ALMR, and systems that utilize thermal neutrons, such as the ATW. Instead, k_∞ was calculated using a spreadsheet. The LWR discharge compositions calculated earlier with ORIGEN2 were reformatted and inserted into a PC-based spreadsheet. Values fork- for each individual TRU nuclide ($k_{\infty,i}$) were calculated using ORIGEN2, and values for $\sigma_{T,i}$ were extracted from ORIGEN2 cross-section libraries for a fast reactor system (such as an ALMR) and a thermal reactor system (such as the ATW). It was then possible to calculate the value of k_∞ within the spreadsheet using the same *method* utilized in ORIGEN2, but without making hundreds of separate calculations with ORIGEN2. In order to accomplish this, Eq. 2 was rewritten in terms $k_{\infty,i}$, as shown in Eq. 3.

$$k_\infty = \Sigma (k_{\infty,i} * \sigma_{T,i} * m_i) / \Sigma (\sigma_{T,i} * m_i). \quad (3)$$

Equation 3 may then be used to determine the values of k_∞ for the mixtures of nuclides of interest (in thii case, the TRU actinides recovered from LWR spent fuel discharged from 1968-2016) due to variations in the initial enrichment, burnup, and cooling time. To simplify, it is assumed that 100% of the TRUS have been recovered from the spent fuel. For the 71,000 metric tons of spent fuel projected to be discharged between the years 1968 and 2016, recovery of 100% of the TRUS provides nearly 800 metric tons of TRUS as fuel for transmutation s ysterns.

RESULTS AND DISCUSSION

Figure 3 shows the variability in k_∞ in a fast reactor for the recovered TRUS from the U.S. inventory of spent fuel (historical plus projected) in 2018, when it is assumed that widespread deployment of transmutation systems as an integral component of the nuclear fuel cycle will occur. It should be noted that the range of k_∞ is from 2.05 to 2.52, even though the majority of the significant (visible) variability

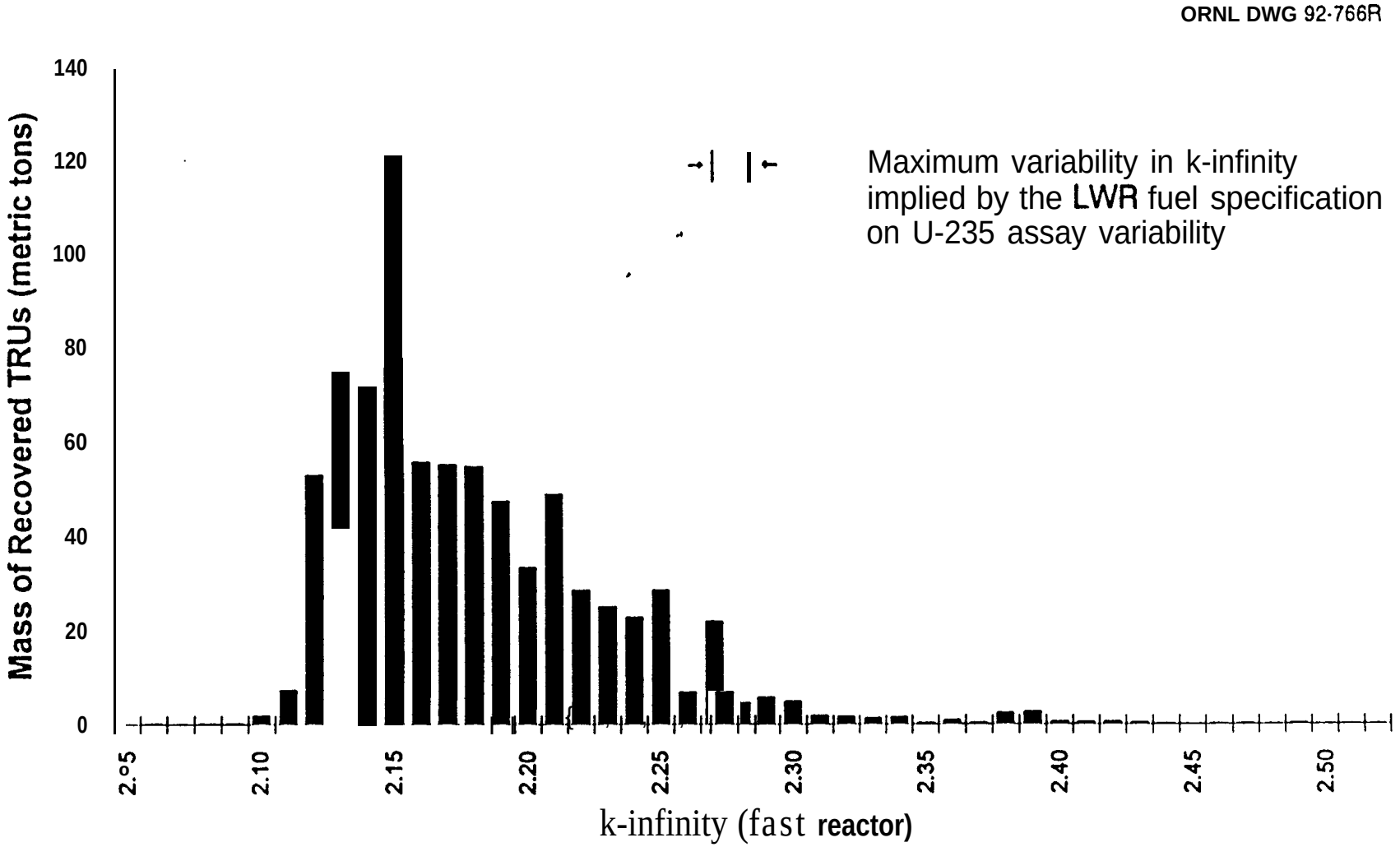

Fig. 3. Variability of fast reactor k-infinity for recovered TRUS for all U.S. LWR spent fuel produced between 1968-2016 by 2018.

is in the range of 2.1 to 2.3. The absolute magnitude of the values of k_∞ should be ignored. Instead, the relative variation in this parameter is simply used as an illustration of the variability in reactivity due to variation in the TRU composition in the spent fuel inventory. The value of k_∞ is determined for an idealized infinite system, and only for the TRUS. We have purposely neglected to include the fission products, structural components, moderator, reactor coolant, reflectors, and other nuclides that would constitute the fuel region of an actual transmutation device.

For partitioning systems, such as pyroprocessing that deal with small batches (i.e., 200 kg of spent fuel containing between 2.5 and 5 kg of TRUS, roughly the same size as a single spent fuel assembly), the full range of variability is important, since partitioning and remanufacture of fuel assemblies using the TRUS as fissile material would be conducted in a batch process, and would not benefit from blending that would occur from other processing options.

To put the k_∞ variability shown in Fig. 3 in perspective, we compare it to the maximum variability in LWR fuel k_∞ permitted by UO_2 fuel specifications. UO_2 fuel specifications[14] require that the ^{235}U assay of fuel be within +-0.05% of the desired assay. In other words, fresh LWR fuel, that may nominally be 3.2% enriched would be acceptable if it falls in the range of 3.15 to 3.25% enrichment. This translates to a maximum variability in k_∞ of 0.015. When compared with the wide variability in k_∞ for the recovered TRUS, it becomes apparent that some means must be developed to assure that fuel for a transmute system can be manufactured to achieve a consistent and fairly narrow range of reactivity constraints, if the transmute has similar requirements. Since transmute system concepts have been developed with the assumption of a fixed isotopic fuel composition, it is important that transmutation systems adequately design for the variation in system reactivity induced by the variability in the TRU isotopic mix.

Figure 4 shows the variability in value of k_∞ in a thermal reactor system, such as the ATW. Again, it is important to note that the range of k_∞ variability k from 1.56 to about 2.10, somewhat wider than the variability in the fast reactor system shown in Fig. 3. One important aspect of a thermal system, such as the ATW, is that such a system is expected to operate at neutron multiplication factors of between 0.90 and 0.95 and be comprised of "cores" (or neutron multiplier blankets) that are nearly 100% TRU elements. As shown in Fig. 4, the variability in k_∞ far exceeds what would be the proposed criticality margin in the accelerator driven cores.

In nonaqueous processing systems, batch sizes are small because high-density forms (e.g., metals) have limits on size due to criticality considerations. Therefore, for these technologies, the processing will have minimal blending and preserve the composition of the source material. Thus, nonaqueous reprocessing technologies have inherent features that make their products more sensitive to the compositional variability in the feed material.

Aqueous processing systems would be expected to have less of batch-to-batch variability since aqueous systems generally involve large volumes of dilute actinides, thus reducing the TRU compositional variability through mixing. Criticality problems are avoided by the dilute nature of the system composition, geometry, and by the addition of neutron poisons. Despite greater blending in aqueous plants, however, some variability in reprocessing plant product will still be expected to exist.

Another method that could reduce the effect of TRU compositional variability would involve the careful measurement of the TRU composition and deliberate batch-to-batch blending so as to meet a specified fissile composition. This method might prove difficult and costly to implement. Aqueous processing, such as a combined PUREX/TRUEX flowsheet affords the opportunity to blend the TRU stream. The traditional PUREX process results in a relatively pure uranium stream, a relatively pure

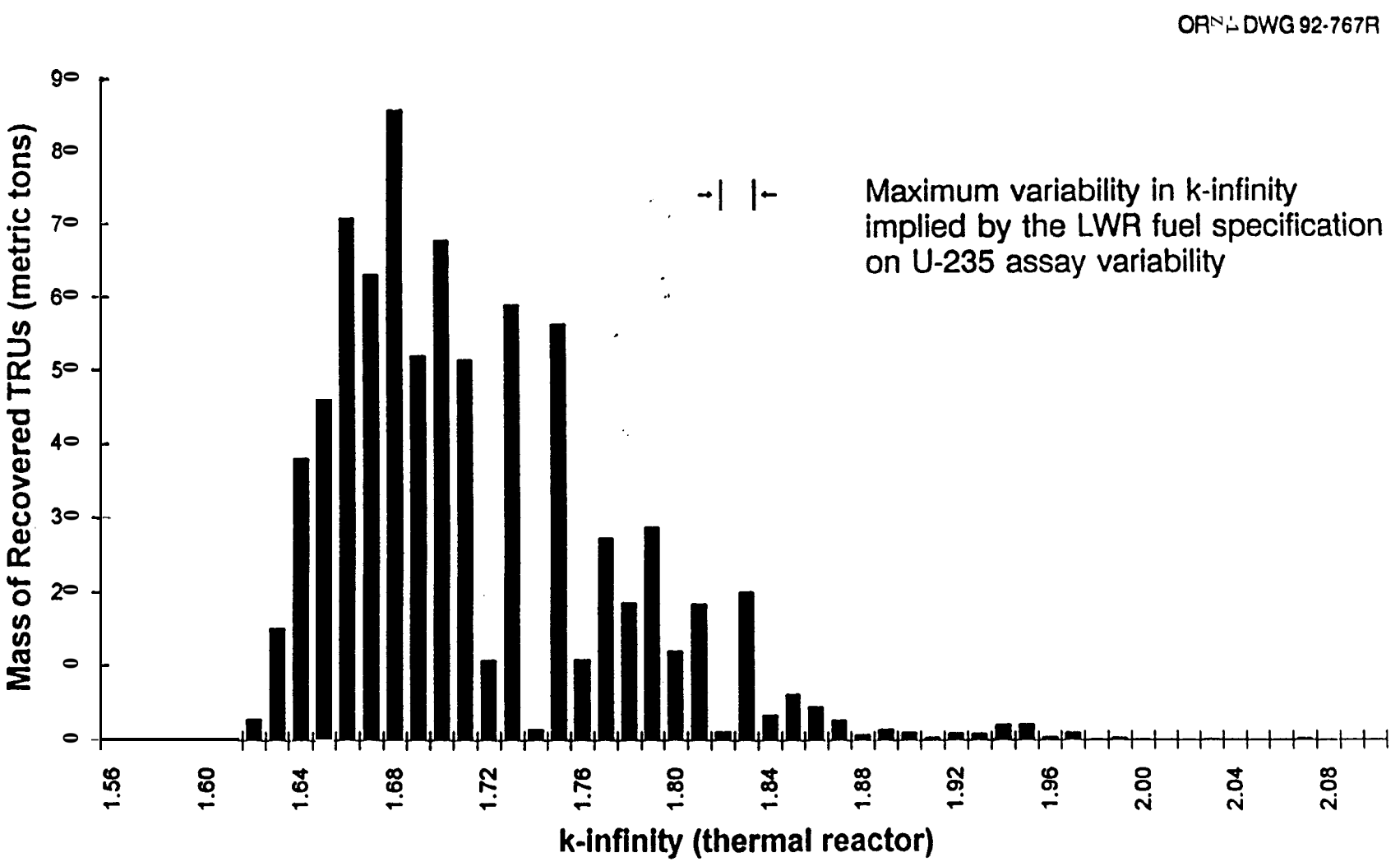

Fig. 4. Variability of thermal reactor k-infinity for recovered TRUs for all U.S. LWR spent fuel produced between 1968-2016 by 2018.

plutonium stream, and a stream containing minor actinides and fission products. Sending the minor actinide/fission product stream through the TRUEX process separates the minor actinides from the fission products. The combined PUREX/TRUEX flowsheet would permit the plutonium and minor actinide streams to be reblended in a controlled fashion, thus achieving a more uniform product. Pyrochemical processing, on the other hand, does not offer an analogous opportunity, since the minor actinide and plutonium streams are never separate during processing.

CONCLUSIONS

The following conclusions may be drawn from this paper.

- Variability in the fuel reactivity in transmute systems, as induced by the variability in TRU isotopic compositions within the U.S. inventory of spent fuel, appears to be significantly greater than the variability in LWR fuel reactivity due to accepted levels of ^{235}U assay variation within LWR fuel.

- The TRU compositional variability appears to be of roughly equal significance with respect to k_∞ in both thermal and fast reactor systems.

- All P-T systems would need to accommodate the variation in TRU composition or specify an acceptable and achievable range. The impact of TRU compositional variability on fuel reactivity in design, licensing, and-operation has not been addressed by P-T system proponents.

- Other aspects of the transmutation system fuel impacted by the TRU compositional variability (e.g., irradiation performance, thermal properties) need to be investigated further.

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