

VALIDATION STUDY FOR CREDITING CHLORINE IN CRITICALITY ANALYSES FOR SPENT NUCLEAR FUEL DISPOSITION¹

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ABSTRACT

Spent nuclear fuel (SNF) management practices in the United States rely on dry storage systems that include both canister- and cask-based systems. The United States Department of Energy Used Fuel Disposition Campaign is examining the feasibility of direct disposal of dual-purpose (storage and transportation) canisters (DPCs) in a geological repository. One of the major technical challenges for direct disposal is the ability to demonstrate the subcriticality of the DPCs loaded with SNF for the repository performance period (e.g., 10,000 years or more) as the DPCs may undergo degradation over time. Specifically, groundwater ingress into the DPC (i.e., flooding) could allow the system to achieve criticality in scenarios where the neutron absorber plates in the DPC basket have degraded. However, as was shown by Banerjee et al., some aqueous species in the groundwater provide noticeable reactivity reduction for these systems [1]. For certain amounts of particular aqueous species (e.g., chlorine, lithium) in the groundwater, subcriticality can be demonstrated even for DPCs with complete degradation of the neutron absorber plates or a degraded fuel basket configuration. It has been demonstrated that chlorine is the leading impurity, as indicated by significant neutron absorption in the water that is available in reasonable quantities for the deep geological repository media under consideration. This paper presents the results of an investigation of the available integral experiments worldwide that could be used to validate DPC disposal criticality evaluations, including credit for chlorine. Due to the small number of applicable critical configurations, validation through traditional trending analysis was not possible. The bias in the eigenvalue of the application systems due only to the chlorine was calculated using TSURFER analysis and found to be on the order of 100 percent mille (1 pcm = 10^{-5} k_{eff}). This study investigated the design of a series of critical configurations with varying amounts of chlorine to address validation gaps. Such integral experiments would support the crediting of the chlorine neutron-absorption properties in groundwater and the demonstration of subcriticality for DPCs in deep geologic repositories with sufficient chlorine availability.

KEYWORDS

Chlorine, Validation, Groundwater, Disposition

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1. INTRODUCTION

The current spent nuclear fuel (SNF) management strategy in the US includes reliance on dry storage systems that include both canister- and cask-based systems to allow continued operation of the nation's nuclear fleet. Approximately 2,000 metric ton of heavy metal (MTHM) is being placed in dry storage per year. This is approximately an additional 200 dry storage casks. An alternative to dual-purpose (storage and transportation) canisters disposal is to repack the SNF into smaller canisters which will significantly increase the cost and worker dose and therefore the US Department of Energy Used Fuel Disposition Campaign is examining the feasibility of directly disposing of these DPCs in a geological repository. Past studies regarding the feasibility of direct disposal have concluded that while possible, demonstrating criticality control over the disposal time period is a challenge. The primary challenge is in demonstrating the continued efficacy of the criticality control features as the system degrades over time (e.g., 10,000 years or more) and groundwater enters the canister. Specifically, groundwater ingress into the DPC (i.e., flooding) could allow the system to achieve criticality in scenarios where the neutron absorber plates between the assemblies in the DPC basket have degraded. However, as was shown by Banerjee et al., some aqueous species in the groundwater may have high enough neutron absorption properties that can be credited to demonstrate subcriticality of the DPC systems over the repository performance period [1]. For certain amounts of amounts of particular aqueous species (e.g., chlorine, lithium), subcriticality can be demonstrated even for DPCs with complete degradation of the neutron absorber plates or a degraded fuel basket configuration. It has been demonstrated that the leading impurity is chlorine, as indicated by significant neutron absorption in the water that is available in reasonable quantity in most of the deep geological repository media under consideration [1]. This paper presents the results of an investigation of the available integral experiments worldwide that could be used to validate DPC disposal criticality evaluations, including the analysis of taking credit for chlorine. Due to the small number of applicable critical configurations, validation through traditional trending analysis was not possible. The bias in the eigenvalue of the application systems due only to the chlorine was calculated using TSURFER analysis and found to be on the order of 100 percent mille ($1 \text{ pcm} = 10^{-5} k_{\text{eff}}$). The current study investigated the design of a series of critical configurations with varying amounts of chlorine to address validation gaps. Such integral experiments would support the crediting of the chlorine neutron-absorption properties in groundwater and the demonstration of subcriticality for DPCs in deep geologic repositories with sufficient chlorine availability.

2. BACKGROUND

Two specific hypothetical configurations were considered as the application models to be covered by the validation study. Both were a 32 SNF assembly capacity DPC models consisting of a stainless steel canister and basket materials with representative 17×17 pressurized water reactor (PWR) SNF. Both cases were modeled with 20 gigawatt-days (GWd) per metric ton of uranium (MTU) burnup as representative burnup for SNF. In the first case, the neutron absorber plate material of the DPC was modeled as moderating material (i.e., infiltrated groundwater) to account for degradation and separation of the absorbing material from between fuel assemblies. In the second case, the fuel basket structure and neutron absorber plates were modeled as moderating material to account for additional potential degradation to the canister basket (Loss of basket materials from between fuel assemblies is considered conservative with respect to criticality evaluations.). An in-depth description and schematics of the two application cases discussed here are available in Ref. 1. The two application models were selected such that the amount of chlorine in the models resulted in a slightly supercritical configuration. The chlorine concentration in the two application models sets a target concentration that would be desirable in the critical experiments used for validation. The ultimate goal of selecting a set of integral experiments is to match the area of applicability relative to DPC systems as close as possible. Table I summarizes the two models.

Chlorine has only two stable isotopes: 75.76% ^{35}Cl and 24.24% ^{37}Cl . The application systems both exhibit a thermal neutron flux. Comparing the thermal capture cross sections of the two isotopes (^{35}Cl : 43.60 b, ^{37}Cl : 0.432 b), it is obvious that only the ^{35}Cl is important for neutron absorption.

Table I. Summary of the two application models

Case	No absorber	Degraded basket
Burnup	20 GWd/MTU	20 GWd/MTU
Cl concentration	25 000 ppm (mg/L)	50 000 ppm (mg/L)
k_{eff}	1.00944	1.05222
Cl worth	0.05113 Δk	0.10365 Δk

The nuclear data library considered in this study is the ENDF/B-VII.1 [3]. The resolved resonance region evaluation for both isotopes of chlorine, which also governs the thermal energy region, was completed in 2003 by an Oak Ridge National Laboratory team led by R. Sayer [4]. The evaluation was subsequently updated in 2007. The goal of the 2003 evaluation was to address several deficiencies in the previous evaluation for chlorine as noted in Ref. 5 in order to support systems where chlorides are present. However, it is important to note that the updated resolved resonance evaluations were never benchmarked on a set of integral experiments.

3. ANALYSIS

3.1 Traditional Trending Analysis

A portion of the computations for this project was done with the SCALE 6.1 code package [6]. In particular, the codes, KENO, TSUNAMI-3D, TSURFER and AMPX were used. A modification of the code SAMINT [7] was used to isolate only the effect of the single chlorine isotope for some of the parameters traditionally computed by TSURFER.

Six critical configurations that could be helpful in validating the capture cross section of chlorine in the thermal energy region were identified as part of the French MIRTE 2.2 program [8]. Of the six configurations, two contain NaCl solution (conc. = 300 g/l), and four have cruciform polyvinyl chloride (PVC) separators in the core. However, these are commercial, proprietary experiments and are not freely available.

The *International Handbook of Evaluated Reactor Physics Benchmark Experiments* [9] was consulted, but no configurations with chlorine sensitivities similar to the two applications were identified. Outside of the *International Handbook of Evaluated Criticality Safety Benchmark Experiments* (IHECSBE) [10], no other source was found that contained potentially applicable evaluated critical experiments with chlorine sensitivities similar to the applications in this research.

A total of 141 critical configurations containing chlorine were identified in the 2013 edition of IHECSBE. Despite the large number of prospective benchmarks, very few have a similar chlorine sensitivity profile shape and magnitude as the application systems of this study.

The sensitivity profiles of k_{eff} for the different chlorine reactions as a function of neutron energy were calculated for the two application models using TSUNAMI-3D from SCALE 6.1. Figure 1 presents the sensitivity profiles for the total cross section of chlorine for the two application systems, as well as for several of the most similar benchmarks.

The HEU-SOL-THERM (HST)-044-003 system is the only benchmark to have a larger sensitivity for chlorine than the degraded fuel basket application system. Notice also that the sensitivity profile of HST-044-003 peaks at a higher energy than the two application systems. While that sensitivity profile has a large magnitude, the shape does not fully resemble that of the two application systems. The LEU-COMP-THERM (LCT)-045-019 benchmark gives an almost perfect match to the no absorber system. Unfortunately, most of the 141 critical benchmarks with chlorine are like HST-008-004 in the sense that they have a very similar shape of the sensitivity profile but a much smaller magnitude. In fact, HST-008-004 is in the top 10 benchmarks when it comes to a quantitative analysis of the similarity between sensitivity profiles.

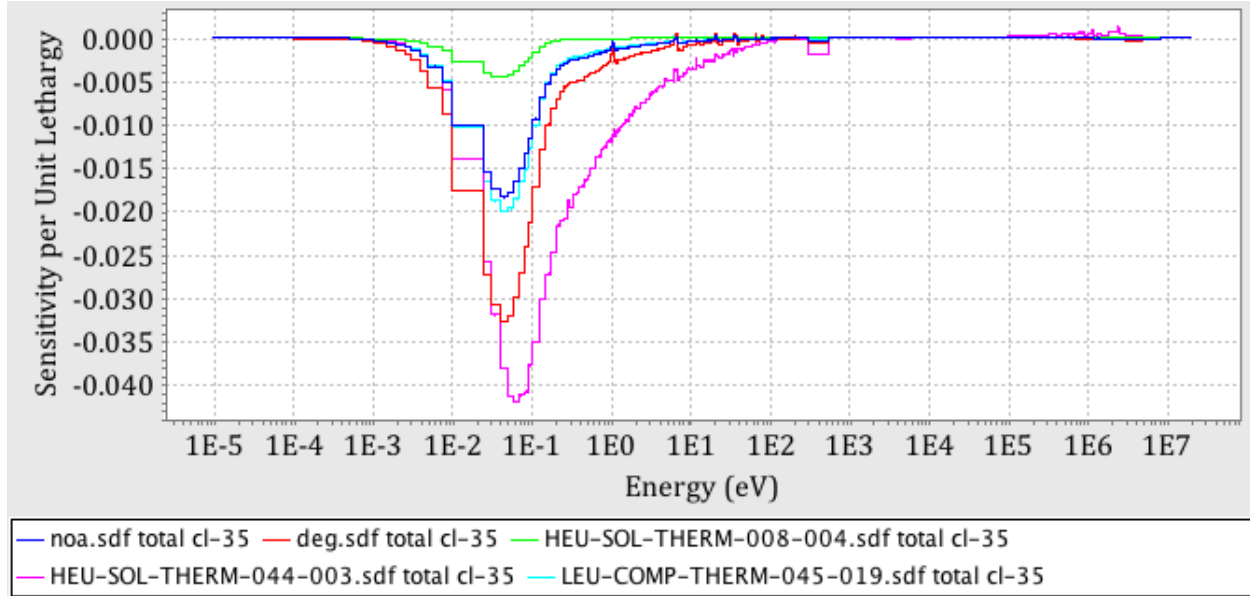


Figure 1. Sensitivity profiles of k_{eff} for total cross section of ^{35}Cl as a function of energy. The two application systems are labeled as *noa.sdf* and *deg.sdf*, which represent the no absorber and the degraded fuel basket systems, respectively.

The 11 most suitable critical configurations are listed in Table II. They originate from four different experiments: LCT45, HST44, HST08, and UST03. For a description and interpretation of similarity coefficients, please see Ref. 2 or Ref. 6. Furthermore, the chlorine content appears as three different materials in the 11 configurations. The chlorine is found in a Plexiglas reflector for the LCT45 and HST08, in PVC rods for HST44, and as a constituent of a paint coating the inside of the solution cylinders in UST03. Based on the chlorine form, it is obvious that none of the experiments have a series of similar configurations where only the chlorine amount changes. All of these factors combine to make validation through traditional trending analysis (regression) difficult. Furthermore, if only the benchmark experiments that have a sensitivity profile for chlorine representative of the application systems are considered, the small sample size results in poor statistics. In this case, neither the normality of the data nor a significantly non-zero trend can be determined.

It is the conclusion of this study that validation through traditional trending analysis is not possible with the current, freely available, evaluated set of critical benchmark experiments. In this case, however, TSURFER analysis is well suited for identifying the level of bias and bias uncertainty based on the available benchmark models.

Table II. Similarity coefficients for the total cross section for the most applicable benchmark experiments compared to the no absorber case

Experiment	G	C	E	Sensitivity
LCT45-18	0.998	0.999	0.999	0.048
LCT45-19	0.998	0.999	0.999	0.048
LCT45-06	0.989	0.999	0.999	0.052
HST44-02	0.916	0.992	0.917	0.066
UST03-02 ^a	0.808	0.999	0.999	0.021
HST44-03	0.740	0.999	0.922	0.135
UST03-04	0.719	0.992	0.999	0.018
UST03-05	0.691	0.999	0.999	0.017
HST08-04	0.488	0.998	0.992	0.010
HST08-12	0.406	0.998	0.991	0.008
LCT45-03	0.401	0.998	0.992	0.008

^a U233-SOL-THERM (UST)-03-02.

3.2 TSURFER Analysis

TSURFER performs a simultaneous adjustment of the cross section data for all of the isotopes within the given covariance data using the generalized linear least-squares approach. TSURFER tries to minimize the cross section changes and the k_{eff} discrepancies for a given set of integral experiments. Since TSURFER adjusts all of the cross section data simultaneously for all of the isotopes, a wide range of integral benchmarks should be used. Alternatively, all of the discrepancy in the k_{eff} could be attributed to an error in a small set of isotopes; in reality, many isotopes contribute to the k_{eff} bias of each integral benchmark. Therefore, the entire set of 394 models in SCALE Verified, Archived Library of Inputs and Data (VALID) [11] was used as the background set of integral experiments to establish the appropriate multigroup cross section changes for all of the isotopes in the two application systems apart from ³⁵Cl. No thermal-neutron-spectrum experiments containing chlorine were part of the VALID library. Two different sets of integral experiments were set up:

- the set of 394 VALID models in addition to the 11 most applicable benchmarks identified in Table II, and
- the set of 394 VALID models and all of the 141 benchmarks that contained chlorine.

The sensitivity data files (SDFs) for the 141 benchmark models that contained chlorine were distributed with the IHECSBE handbook. These models are not considered to be as reliable as the VALID models. While the benchmark evaluations in the IHECSBE handbook undergo a rigorous review process, neither the computational model inputs nor the SDF files distributed with the handbook are subjected to a review process as part of the IHECSBE effort. In contrast to the IHECSBE, the SCALE VALID process was set up to ensure high quality model input and SDF files through a review process. However, the 11 most applicable benchmark models identified in Table II were hand checked, and the calculated chlorine sensitivity was verified by direct perturbation calculations. The difference between the two sets of the experiments mentioned above is that the first set could be considered reliable, but with the scope of the chlorine-containing benchmarks limited only to the most applicable ones, and the second should not be considered reliable but encompassing all of the freely available data. Bias is defined as the systematic difference between calculated results and experimental data. Note that following the convention of bias, a positive bias for the chlorine is a conservative bias with respect to the safety analysis case. In other words, a positive bias associated with chlorine may indicate that, based on the TSURFER analysis, the

applications' calculations are higher because of an error in the chlorine cross sections. This may suggest that the chlorine capture cross section in the current ENDF/B-VII.1 evaluation could be increased slightly.

The propagated chlorine uncertainty was calculated using the SAMINT code. It is evident from Table III that the exact numbers for the calculated bias and bias uncertainty depend on which set of benchmark experiments is used in the analysis. However, the same pattern emerges regardless of the set of integral experiments used. The propagated k_{eff} uncertainty from all of the isotopes for both application systems is around 550 pcm. The ^{35}Cl uncertainty contributes approximately 50 pcm uncertainty to the k_{eff} of the no absorber application case and 100 pcm to the k_{eff} uncertainty of the degraded fuel basket case. In all cases, both the bias from all of the nuclear data and the bias just from the ^{35}Cl are less than the calculated uncertainty. Furthermore, it is clear that the uncertainty in the chlorine cross section can be considered to bound the bias. A similar argument has been previously made for fission product isotopes that had very limited or no critical experiments available [12].

Table III. TSURFER results

	No absorber	Degraded basket
Initial k_{eff}	1.00940 +/- 0.00544	1.05220 +/- 0.00552
238 group propagated Cl initial uncertainty ^a	0.00058	0.00109
44 group propagated Cl initial uncertainty	0.00056	0.00102
<i>Using all VALID benchmarks and the 11 most applicable chlorine containing benchmarks using ENDF/B-VII.1 covariance data for ^{35}Cl with a flat flux collapse</i>		
Total bias	-0.00127	-0.00066
Final k_{eff}	1.01070 +/- 0.00148	1.05290 +/- 0.00144
^{35}Cl bias	0.00037	0.00070
44 group propagated Cl final uncertainty	0.00052	0.00094
<i>Using all VALID benchmarks and the 141 chlorine containing benchmarks using ENDF/B-VII.1 covariance data for ^{35}Cl with a flat flux collapse</i>		
Total bias	-0.00016	0.00032
Final k_{eff}	1.00960 +/- 0.00110	1.05190 +/- 0.00141
^{35}Cl bias	0.00021	0.00040
44 group propagated Cl final uncertainty	0.00053	0.00096

^a One standard deviation is presented as a measure of uncertainty.

4. NEW EXPERIMENT DESIGN

The current study investigated the design of a series of critical configurations with varying amounts of chlorine to address the validation needs. Such integral experiments would support the crediting of the chlorine neutron-absorption properties in groundwater and the demonstration of subcriticality for DPCs in deep geologic repositories.

Within the US, the Burnup Credit Critical Experiment (BUCCX) at Sandia National Laboratory is the most likely facility to produce a series of critical experiments that could be used for a traditional validation study through trending analysis. The International Criticality Safety Benchmark Evaluation Program (ICSBEP) experiment LEU-COMP-THERM-079 was taken as the basis for a study of the

possible modifications to the experiment such that the bias and uncertainty due to chlorine could be identified.

The original LEU-COMP-THERM-079 was an experiment designed to isolate the effects of rhodium. In a hypothetical situation, the Rh foils in experiment LCT79-03 were simply replaced with chlorine. Figure 2 presents the resulting sensitivity profile compared to the no absorber application.

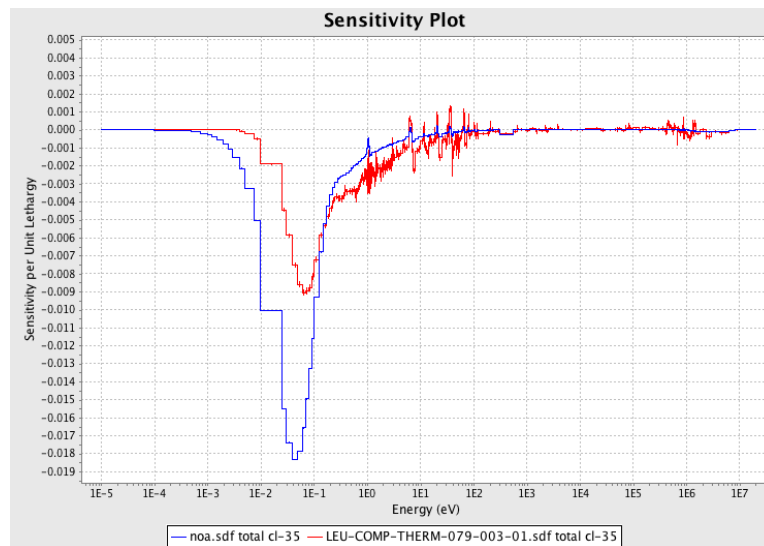


Figure 2. Sensitivity profile for the total cross section of chlorine for a modified version of the LCT79-03 configuration and the no absorber application.

This important figure suggests that fairly large magnitudes of sensitivity to chlorine can be achieved. More importantly, Figure 2 suggests that the chlorine-containing material must be separated from the fuel by some amount of moderation in order to achieve a peak in the sensitivity profile at lower neutron energy.

As a further test, the rhodium was removed from two of the LCT79 models, and the same concentration of chlorine was added to the water in the model as was used for the no absorber application (25,000 ppm). It is not proposed to flood the BUCCX experiment with chlorinated water; rather, this is a demonstration calculation to show the achievable similarity in the chlorine sensitivity profile. Figure 3 presents the resulting sensitivity profile.

The modified LCT79-5 and LCT79-10 experiments differ in the pitch of the lattice and also in the number of fuel rods necessary to make each critical when the rhodium was present. Figure 3 shows an almost ideal match for the chlorine sensitivities. If the same chlorine worth can be efficiently distributed throughout the moderator, a very high similarity coefficient for chlorine can be expected.

The simplest way to distribute the chlorine throughout the LCT79 core is to place a chlorine solution within dummy fuel rods and evenly spread them out throughout the lattice. The optimization goal in this design is to get the maximum possible chlorine worth configuration. The two different pitches for the LCT79-5 and LCT79-10 were 2 cm and 2.8 cm respectively, with the latter configuration considered almost optimally moderated. The 2 cm pitch gives an areal rod density of nearly double that of the optimally moderated lattice. Therefore, to achieve near maximum chlorine worth with dummy chlorine

rods, the configuration shown in Figure 4 is proposed. The design is to replace every third fuel rod of the LCT79-5 configuration with a chlorine dummy rod. This achieves a uniform distribution of chlorine throughout the moderator and results in a near critical configuration with all of the lattice positions filled when the maximum chlorine concentration is used in the dummy rods.

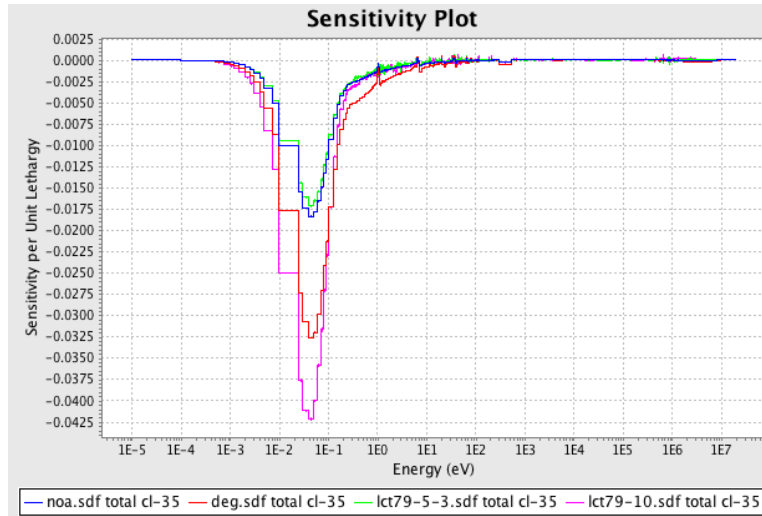


Figure 3. Sensitivity profile for the total cross section of chlorine for two modified version of the LCT79 configuration and the two application cases.

The proposed design involves 264 fuel rods, 1 source rod and 132 chlorine dummy rods. When the chlorine dummy rods are filled with a salt-water solution of maximum sodium chloride concentration of 350 ppm, the calculated system k_{eff} is 0.988605 ± 0.00010 . The calculated chlorine worth for this system is approximately 9,600 pcm. Figure 5 presents the calculated k_{eff} values for a series of configurations with varying chlorine concentrations.

Figure 5 shows that the proposed experimental set up responds linearly with respect to the chlorine concentration in the dummy rods. Figure 6, presents the corresponding sensitivity profiles compared against the two application cases. Further, Table V reports the calculated similarity coefficients for only the ^{35}Cl and for all of the isotopes.

The total similarity coefficients are relatively small between the proposed experimental series and the applications despite similar neutron energy spectra, lattice configuration and moderating material. The disagreement is due to the difference in the fuel. The application fuel being 20 GWD/MTU used nuclear fuel compared to fresh fuel used in the critical experiments. However, with respect to the chlorine similarity coefficients, Table V quantifies what is evident from Figure 6. The proposed series of critical configurations has a very high degree of similarity with respect to the sensitivity profile for ^{35}Cl . Furthermore, the series of experiments is able to achieve the necessary magnitude of the sensitivity profile to match each of the application cases.

In this preliminary study, it has been shown through Monte Carlo simulations, that a plausible set of critical configurations could be assembled at the BUCCX at Sandia National Laboratory that would be able to fill in the gap in the lack of critical benchmark experiments with chlorine sensitivity in the thermal energy region. While the proposed experiment does not match with a high degree of similarity the two application cases on an overall-nuclide basis, the sensitivity profiles for chlorine are predicted to be almost identical as the two application cases. Such a series of critical experiments would be invaluable to

providing additional confirmation that nuclear data uncertainty can be used to bound the bias for unvalidated nuclides (e.g., chlorine) in criticality safety applications as discussed in Ref. 12.

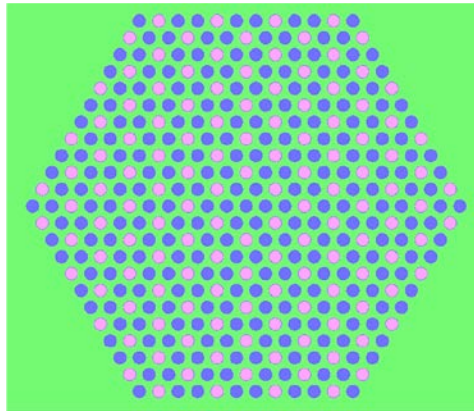


Figure 4. Proposed core configuration for modified version LCT79-5. Fuel rods are blue; chlorine dummy rods are pink.

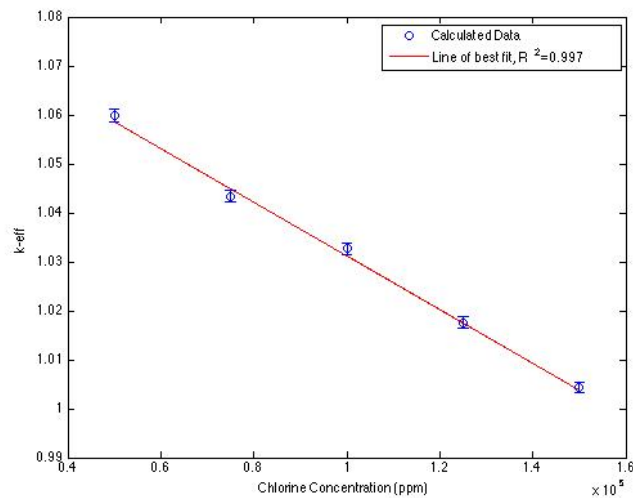


Figure 5. k_{eff} values for a series of 5 configurations with varying chlorine concentration.

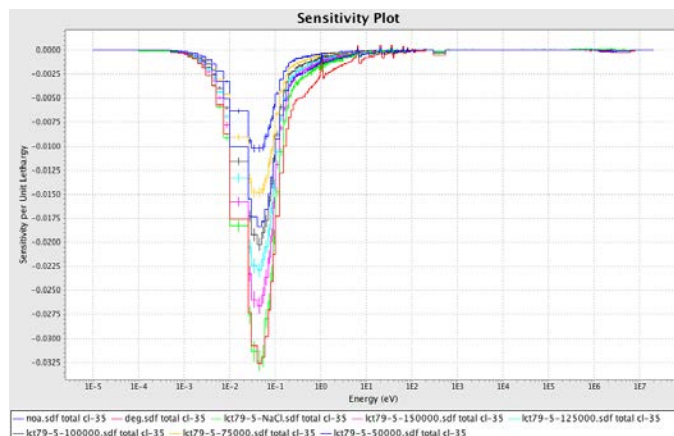


Figure 6. Sensitivity profiles for ^{35}Cl total cross section of proposed experimental series. File names include chlorine concentration value (ppm). The configuration labeled lct79-5-NaCl.sdf has maximum dissolvable salt (NaCl) concentration in dummy rods.

Table V. Similarity coefficients for the proposed experimental series.

Application	Similarity for ^{35}Cl						Total Similarity	
	No absorber			Degraded basket			Noa	Deg
Chlorine content	C	E	G	C	E	G	C	C
Saturated NaCl	0.9998	0.9992	0.8571	0.9996	0.9983	0.9983	0.467	0.494
150 000 ppm	0.9997	0.9983	0.9178	0.9994	0.9971	0.9854	0.463	0.488
125 000 ppm	0.9996	0.9987	0.9634	0.9994	0.9976	0.9544	0.457	0.479
100 000 ppm	0.9995	0.9982	0.9883	0.9993	0.9970	0.9120	0.462	0.483
75 000 ppm	0.9995	0.9975	0.9916	0.9992	0.9960	0.8036	0.461	0.480
50 000 ppm	0.9993	0.9968	0.9021	0.9991	0.9952	0.6330	0.465	0.483

5. CONCLUSIONS

The available literature was surveyed for evaluated critical benchmark experiments that could be used in a validation study to support crediting chlorine as part of criticality analyses for SNF disposal. Two particular DPC models were considered as application models. Both DPC application models were assumed to be flooded by groundwater containing chlorine, with one model having the absorber plate completely deteriorated and the other having the fuel basket and absorber plates completely disintegrated. From the noncommercial resources, only the IHECSBE was found to have evaluated critical experiments with chlorine sensitivities similar to the two application cases. A total of 141 integral experiments were identified to contain chlorine; however, only 11 of these had near enough sensitivity to be considered as a suitable representation of the chlorine in the application systems. Therefore, traditional validation of chlorine through trending analysis was deemed impossible due to the small number of relevant experiments and their diverse nature.

The code TSURFER provided an estimate of the bias uncertainty for the application systems. The bias uncertainty is estimated at a one-sigma level to be around 50 pcm for the no absorber application case and around 100 pcm for the degraded fuel basket case. Currently there are no set criticality safety criteria for licensing of SNF disposal. Assuming that possible future criteria will be similar to those used for spent fuel pools, based on current regulation as prescribed in Reference 13, quantification at the 95% probability with 95% confidence upper subcritical limit (USL) will need to be calculated. However, there currently is no known way to meet the above requirement as written. There is a clearly defined way to calculate a USL from the TSURFER analysis; however, it is not clear how the USL determined with the TSURFER procedure will meet the 95/95 requirement. A dedicated set of critical experiments similar to the proposed application would allow for a clear determination of the 95/95 USL using traditional trending analysis. One possible experiment was modeled and determined to be highly likely to fill the chlorine gap in the experimental database. Furthermore, trending analysis conducted on a dedicated set of integral experiments would support and validate the TSURFER analysis.

Recommended next steps would be to automate the procedure described in this report for the validation study for crediting chlorine in criticality analyses for SNF disposition to a general procedure that can be used for any isolated chemical element. Such an automated procedure would be a useful extension to the Used Nuclear Fuel Storage, Transportation & Disposal Analysis Resource and Data System (UNF-ST&DARDS) [14] being developed at Oak Ridge National Laboratory as a collaboration between several national laboratories and industry partners. UNF-ST&DARDS has an automated procedure that assembles

as-loaded criticality models for desired DPC configurations. From that point, a general procedure could be constructed that would execute a TSUNAMI-3D run followed by TSUNAMI-IP and TSURFER calculations. The end results would be the uncertainty in k_{eff} associated with the targeted nuclide cross sections, and the bias and bias uncertainty according to the TSURFER calculation for the application as a whole and just for the individual chemical element of interest. This information would support future licensing efforts.

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