Title: Discussion of NRC FQ/MST Assessment Report Follow Up Items Related to Neutron Flux Spectrum and Effects of Silver, Palladium and Neutron Flux on Radionuclide Transport through Silicon Carbide

TEV No.: 1620 Rev. No.: 0 Project No.: 23841 Date: 09/10/2012

# Technical Evaluation Study

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## INTRODUCTION

The Nuclear Regulatory Commission staff, in its review of the NGNP Fuel Qualification and Mechanistic Source Terms White Papers, has raised a number of follow up items regarding the prototypicality of coated particle fuel irradiation testing in the Advanced Test Reactor (ATR) for the AGR Fuel Development and Qualification program. The items raised pertain to the differences in neutron flux spectrum between the ATR and gas-cooled reactors, differences in plutonium fission rates and the resulting production rate of fission products such as silver and palladium, and the possible differing effects of Ag, Pd, and neutron flux on the diffusion of other fission products (e.g., cesium) through the SiC layer in TRISO coated particle fuel. These follow up items were documented in the NRC Assessment Reports on the white papers and are listed in Attachment 1, which also provides a summary of NGNP/NRC discussions to date on these follow up items.

This paper documents the AGR Program's current evaluations of these items based on an assessment of the historical database and results from the AGR-1 PIE to date. In its review and in discussions with the AGR Program staff, NRC has suggested some changes to irradiation test design that it believes would result in a test environment that is more prototypic of a gas-cooled reactor. These potential changes are discussed, and the program's assessment of the suggestions is provided in this document.

## ADDITIONAL INFORMATION

### Ag, Pd, and neutron flux effects on diffusion through SiC

The NRC staff has raised follow up items related to the effects of silver, palladium and neutron flux on the retention of fission products such as cesium by the SiC layer of the TRISO coating. The items include: (a) the effects of large amounts of Ag diffusing through the SiC at high fuel irradiation temperatures (>1200°C) potentially changing the retentiveness of the SiC layer, (b) large amounts of Pd produced from high burnup LEU TRISO fuel potentially attacking the SiC, and (c) neutron flux increasing the diffusivity of cesium or other fission products through the SiC layer. The staff is concerned that these phenomena could all result in the SiC coating layer being less retentive of fission products than expected by the NGNP program.

The following information is offered relative to these hypotheses. This information forms the basis for the AGR program strategy at this point in time.

1. AGR-1 Irradiated Microsphere Gamma Analysis (IMGA) during PIE (at INL and ORNL) is demonstrating significant release of Ag from fuel particles in a compact but no detectable Cs release. If Ag release were enhancing the transport of cesium through the SiC layer, one would expect to have seen cesium in the deconsolidated matrix material or outside of the compact on the capsule components. Nothing beyond that expected from the presence of heavy metal contamination in the matrix was observed. In the one compact where an exposed kernel was found, cesium was found in amounts equivalent to one particle inventory. Thus, there is no evidence in PIE results to date that Ag diffusion through the SiC coating has any effect on the diffusivity of other fission products through the coating.

2. The Cs IMGA results and the minimal cesium in the fuel matrix as measured by deconsolidation-leach-burn-leach analysis suggest there was no Pd attack that degraded the SiC layer to the point of affecting cesium retention in the five compacts examined thus far. (The five compacts span a range of burnups from 11 to 19% FIMA.) Thus, while Pd interaction with the SiC could possibly enhance the release of other fission products, this effect has not yet been observed in AGR-1 PIE.

3. In the three compacts examined to date for which a measurement of stable Pd isotopes was performed, about 1% of the Pd is found in the matrix outside of the SiC, yet no Cs release has been observed. Clearly Pd transported through the SiC layer, but detailed microstructural analysis has shown no evidence of Pd attack of the SiC. Instead, Pd is found in small clusters or precipitates at the SiC/IPyC interface and in the SiC. PIE from EU-1bis has similarly found Pd in German fuel in the IPyC layer where the SiC penetrated the porosity of the IPyC during SiC layer coating. The AGR-1 PIE continues to better characterize the observed behavior.

4. The historic German data indicate that Cs release in heating tests at low temperature (1200°C) is consistent with the QC data on exposed kernels as noted in the NGNP Fuel Qualification White Paper. This implies that there was no enhanced cesium release because of Ag or Pd in the German irradiations.

5. In AGR-1, no cesium release via diffusion has been observed thus far in PIE. If one calculates a breakthrough time, assuming t is the irradiation time and a is the thickness of the SiC layer, a maximum estimate of diffusion coefficient $D = a^2/6t = (35E-06)^2/(6*620\ day*24\ hr*3600\ s) = 3.8E-18\ m^2/s$ is obtained. This value is very close to that recommended in "Fuel Performance and Fission Product Behaviour in Gas Cooled Reactors," IAEA TECDOC 978, November 1997.

6. In the limited heating test data to date, Cs release at high temperature (1600 or 1700°C) is the result of exposed kernels or a SiC failure. Little diffusive Cs release has been observed after three hundred hours of heating in several compacts.

7. Similar data will be obtained from the hot capsule (1400°C) in AGR-2 (and capsules in AGR-7) that will provide information on in-pile diffusional release at higher temperature irradiation. If some cesium is seen in the matrix, the diffusion coefficient in the SiC can be established based on a transient diffusion solution in PARFUME. The Cs in the matrix from a compact in the hot capsule can also be compared with the Cs in the matrix from a compact irradiated at lower temperature and safety tested at 1400°C. This may provide a more direct comparison of the effects of irradiation. In this context, the NRC staff expressed a concern that the German data on fission product diffusion through SiC from IAEA TECDOC 978 are based largely on integral safety testing. For clarification, the values that are recommended in the TECDOC are based on a combination of historic irradiation testing from BISO particles for PyC, loose kernels for the kernel, and some separate effects irradiation testing, as well as post-irradiation integral heating test data. The recommended model for Cs diffusion through SiC in the TECDOC includes an effect of neutron fluence.

8. Based on the best available knowledge, Pd attack is a much stronger function of temperature than of concentration (burnup). Japanese work [K. Minato, T. Ogawa, S. Kashimura, K. Fukuda, M. Shimizu, Y. Tayama, and I. Takahashi, "Fission Product Palladium-Silicon Carbide Interaction in HTGR Fuel Particles," J. Nucl. Mater. 172, 184 (1990).] suggests very weak burnup dependence, albeit at very low burnups. Minato

suggests that SiC corrosion is proportional to the concentration of Pd (burnup) to the 1/3rd power. This is a very weak dependence.

9. Pd attack has historically only been observed in US and Japanese fuel irradiations. None was found in German irradiations. This may be, in part, because US and Japanese irradiations prior to AGR were very accelerated; thus kernel temperatures could be much higher than the temperature of the fuel matrix.

10. In the FTE-13 experiment, $PuO_{1.68}$ was irradiated to 70% FIMA for 900 EFPD at peak temperatures between 1146 and 1444°C (average temperatures between 864 to 1031°C), depending on the capsule, and fast fluences between 2 and 2.26 x $10^{21}$ n/cm$^2$. Some Pd interaction with SiC was observed, but no large-scale degradation of the SiC layer was observed. Calculations suggest that the volumetric Pd concentration in the FTE-13 kernels was 75x that in AGR-1 kernels. The areal (surface) Pd concentration (total number of Pd atoms divided by geometric surface area of a kernel) in FTE-13, a metric for mass flux across a surface, is 60x that of AGR-1. The FTE-13 irradiations of pure plutonium fuel substantially envelope the plutonium fission rates expected for the NGNP and provide confidence that the phenomena raised in the NRC follow up items will not be significant under NGNP irradiation conditions.

11. As much of the literature suggests, Pd corrosion is controlled by release from the kernel. Simple out of pile diffusion couples suggest that if large quantities of Pd transport to the SiC, the penetration rates into the SiC are very fast, substantially higher than observed in PIE of TRISO fuel. This result highlights the shortcomings of out-of-pile experiments that attempt to simulate what occurs in TRISO fuel, which is more complex. Out-of-pile experiments and simple model systems need to establish the proper chemical activity of the Pd to directly compare with actual TRISO fuel. Based on the current understanding of fission product release from the kernel (and from $UO_2$ fuel pellets in LWRs), metallic fission products like Pd, Mo, Tc, Rh, and Ru tend to form metallic inclusions in the fuel because of their immiscibility in the ceramic phase. Thus, there is an aggregation of nodules that form in the fuel with increasing burnup. Release of Pd from the fuel kernel through the buffer and inner PyC to the SiC layer is then determined by vapor transport from these nodules, which is strongly influenced by the temperature of the kernel and the partial pressure of the Pd in the nodule (i.e. vaporization from a metallic solid solution). This picture of release would not be expected to show a strong dependence on burnup or concentration.

12. The AGR irradiations will produce smaller inventories of Ag and Pd than in an HTGR because of differences in the neutron flux spectrum. However, the neutron flux in AGR irradiations is greater than that in an HTGR and is thus conservative regarding its possible influence on diffusivities through the SiC.

13. Because AGR irradiations are accelerated, the irradiation time is shorter in the AGR irradiations than in an HTGR. Thus, the time at temperature is less than in an HTGR, which could curtail diffusion distances and potential releases through the SiC during irradiation. On the other hand, the higher neutron fluxes associated with accelerated irradiations tend to steepen the radial temperature gradient through the fuel particles, increasing kernel temperatures, which may have a larger effect on diffusive releases than longer times at lower temperatures. Analysis of the accelerated effects of irradiation [as discussed in more detail in J. T. Maki et al., "The challenges associated with high burnup, high temperature and accelerated irradiation for TRISO-coated particle fuel," Journal of Nuclear Materials, Vol.

371 (2007) pp. 270–280] suggest that the dominant mechanism depends on the fission product of interest and the physical dimensions of the particle, but for the TRISO particle being qualified by NGNP the effects on the diffusion of Ag and Cs, and on the penetration of SiC by Pd are not very large and are well within the current uncertainty of the experimental database.

Thus, it is not clear that the NRC hypotheses about the deleterious effects of Ag, Pd, and neutron flux on transport of Cs through SiC are correct. All the data have not yet been obtained from AGR-1, but no data have been found that support the stated concerns. Of course, if data from the PIE of AGR-1 provide new insights, they will be accounted for in the AGR program going forward.

**Spectral effects of ATR vs. HTGR and their impact on AGR irradiation testing**

The concentration of Ag produced from ATR testing is about 40% below that expected in a prismatic HTGR at a peak burnup of ~20% FIMA because of the differences in the epithermal parts of the neutron spectrum and the differences in plutonium fission rates. Overall these differences are believed by the AGR Program to be small in comparison with the uncertainties in the phenomenology of Pd interaction with the SiC coating, the weak concentration dependence of the interaction (based on today's understanding), and the much larger effect of temperature on the interaction compared to concentration.

NRC staff has provided three suggestions to increase plutonium fissions and generate more Ag and Pd. These suggestions have been discussed in meetings between the AGR Program staff and the NRC, and the AGR Program has evaluated the feasibility of implementing each suggestion. This evaluation has indicated that each suggested approach requires a different balance among competing considerations for AGR test design, data collection, and program execution. The following briefly summarizes the program's assessment of each suggested alternative:

1. Using a Lower Uranium Enrichment: A lower enrichment would generate more Pu fissions from U-238 capture. Significant reductions in enrichment levels will probably be required to obtain a representative plutonium fission rate. However, significant reductions in enrichment while reaching the same burnup would also reduce the heat source in the capsule due to U-235 depletion during the irradiation and would make it impossible to hold temperature at the target burnups the program is trying to achieve.
2. Using Thermal Neutron Shielding to Harden the Neutron Spectrum: This was done in AGR-1 and AGR-2 through a combination of Hf and borated graphite. However, within the limitation of test capsule design, it does not modify the spectrum enough to obtain a representative plutonium fission rate. Calculations on the spectral effects in the ATR Northeast Flux Trap are underway to see whether further modification of the neutron spectrum through use of shielding is possible at this location in the ATR core for AGR-5/6/7.
3. Using Pu in the Fuel Kernels: This approach presents substantial issues relative to current licensed coated particle fuel fabrication capability. Currently B&W does not have a license to handle Pu, and there is no other coated particle fuel fabrication capability in the U.S. licensed to handle Pu in the quantities needed. Changing the license for B&W would be a

very lengthy and expensive effort, if it could be done. In addition, the AGR 5/6/7 irradiation is being used to qualify the TRISO fuel fabrication process, which is a very high priority of the AGR Program. Fabrication of plutonium-bearing coated particle fuel would not contribute to this priority.

## SUMMARY

Based on the assessment of the historical database and results from AGR-1 PIE obtained to date, there is no evidence to support the NRC hypotheses about the deleterious effects of Ag, Pd and neutron flux on transport of Cs through SiC. As the PIE of AGR-1 continues, if the data provide new insights, they will be accounted for in the AGR program going forward.

Based on the considerations addressed in this paper, there is limited value in more closely simulating the exact HTGR neutron spectrum and plutonium fission rate, and there is insufficient flexibility to modify test conditions to more closely simulate the exact HTGR conditions. This conclusion will continue to be re-visited as PIE of AGR-1 continues and the design of AGR-5/6/7 commences.

## ATTACHMENT 1

## NRC FQ/MST Assessment Report Follow Up Items Related to Neutron Flux Spectrum and Effects of Silver, Palladium and Neutron Flux on Radionuclide Diffusion through Silicon Carbide

### FQ/MST-2, Additional Fuel Operating Condition Parameters

*"Fuel service conditions for normal operations should be supplemented with parameters that address maximum plutonium burnup (fissions from bred plutonium) and maximum time-at-temperature."*

### FQ/MST-12, Prototypical Irradiation Testing Neutron Spectrum

*"The NGNP approach to increasing plutonium burnup in the AGR irradiation tests relies solely on using neutron absorbers in the test rig to effectively harden the thermal spectrum by reducing the neutron flux in the lower range of the ATR thermal energy spectrum. The working group presently views this approach as unlikely to adequately address plutonium burnup, time at operating temperature, and particularly palladium time-at-temperature, important parameters that should be considered in the irradiation testing of TRISO fuel."*

### FQ/MST-13, Palladium, Silver, and Rare Earth Time at Temperature

*"Plutonium burnup, time at operating temperature, and particularly palladium time-at-temperature are important parameters that should be considered in the irradiation testing of TRISO fuel."*

### Summary of NGNP/NRC Discussions to Date:

4/17/12 NGNP/NRC Meeting: NRC clarified that plutonium burnup would be measured in terms of Pu fissions per initial heavy metal atom (i.e., per initial uranium atom). The possibility of mixing some lower enriched particles with higher enriched particles in AGR-5/6 to obtain higher plutonium burnup was discussed.

NRC is concerned about non-uniform presence of Pd on SiC (inclusions). Would be interested in the results of a long term postirradiation anneal, perhaps on FTE-13 particles. Use of Pd/SiC diffusion couples was also discussed. NRC agreed that effects of rare earths are not a concern based on NGNP responses to RAIs.

NGNP will consider how it can provide the information of interest and will follow up with NRC. Some of the information of interest will become available as the AGR fuel program proceeds, including measurements of the rate of palladium penetration into the SiC coating.

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**FQ/MST-21, Flux-Accelerated Diffusion of Metallic Fission Products During Irradiation**

*"For the AGR-2 and AGR-7 tests, the Project states that the release of fission products under irradiation will be measured via PIE, that these measurements will be analyzed to establish diffusion coefficients under irradiation, and that the resulting diffusion coefficients will be compared to the historic values from IAEA-TECDOC-978. NRC staff evaluation of the results of these planned actions is a follow-up item."*

**Summary of NGNP/NRC Discussions to Date:**

4/17/12 NGNP/NRC Meeting: FQ/MST-NRC summarized this topic and acknowledged that it is a theoretical concern at this point. NGNP stated that no cesium release has been measured so far in PIE of AGR-1. NRC will continue to evaluate this topic as additional AGR data become available, but no further near term action is required.

NRC Assessment Report
Follow Item FQ/MST-38
*Uncertainty Models*

***The working group notes that the Monte Carlo uncertainty analysis proposed by the Project appears to address only parametric uncertainty. The regulatory community recognizes also "model uncertainty" and "completeness uncertainty". There is, of course, no practical way to quantify completeness uncertainty ("unknown unknowns"). There is, however, a growing trend of asking at least for some assessment of model uncertainty if not rigorous quantification of this uncertainty. This is an item for follow up.***

Follow up item FQ/MST-38 was discussed at the April 17, 2012, NGNP meeting at the NRC headquarters offices. The staff clarified that they did not intend that "completeness uncertainty" be addressed explicitly. However, they were interested in the NGNP Project's approach to "model uncertainty." In response to that request, the following outlines the project's plans.

**Paraphrased Follow-Up Question: How is the NGNP Project going to treat model uncertainty as opposed to parametric uncertainty in the MST and resulting dose uncertainty analyses?**

**Response:**

The mechanistic source terms (MST) and resulting dose uncertainty analyses include assessments of the plant response to transients and off-normal events, MSTs, and the resulting offsite radiological consequences:

In a Level 1 PRA, the separation of uncertainties into model and parametric uncertainties in the frequency of event sequences is commonplace. In principle, this division of uncertainties can be applied to the MST consequences as well.

In a Level 1 PRA the parametric uncertainties are largely associated with the parameters of reliability-type models for initiating events, equipment failure rates, maintenance unavailabilities, human error probabilities, and common cause failure model parameters. Hence, the parameters themselves are parameters of models so they are not really distinct from modeling assumptions, and the idea that they are separate is somewhat artificial. In Level 1 PRA there are selected areas where additional modeling uncertainties are incorporated. (LWR examples include models for reactor coolant pump seal LOCAs, success criteria assumptions, etc.) The methods for quantifying both parametric uncertainties and modeling uncertainties include use of Bayes' methods for incorporating evidence for different sources as well as expert elicitation and opinion.

In the analysis of source terms and accident phenomena there are also roles for both parametric and modeling uncertainty treatment, but the nature of the parameters and models differs from that normally addressed in a Level 1 PRA. Consequence models are ideally rooted in the laws of physics, thermodynamics, chemistry, etc. The "parameters" that are subject to uncertainty may be limited to well defined variables such as material properties, heat transfer coefficients, etc. However, in practice the laws of physics that control the relevant processes may not be fully understood, in which case parameters are often used to model uncertainty in the underlying

models. Examples of these parameters include fractions of radionuclides involved in various radionuclide transport processes. Hence there is great difficulty achieving a clean separation between model and parametric uncertainties.

Although there are different physical phenomena at play, the approach to treatment of consequence uncertainties for the HTGR is very similar to the approach for LWRs discussed in NUREG-1150:

a. When two or more different computer codes or models are available to model the same phenomenon, each is utilized and probabilistically weighted.

b. In some cases parametric models are adopted in lieu of physical models, such as on fractions of radionuclides involved in various processes, which are supported by calculations and/or expert judgment.

c. Formal expert elicitation may be used to address certain aspects not well represented by physical models (as was done for the PBMR NGNP retention parameters and for the NUREG-1150 source terms).

d. Use of expert judgment will follow the requirements of the PRA standard (the draft non-LWR standard uses the same requirements as the current LWR standard for this aspect); those requirements reference NUREGs that have rules for when different models are used versus only assigning a distribution to a parameter.

e. From a computational perspective both probabilistic Monte Carlo and sensitivity analyses are used to combine the uncertainties into the consequence results.

Although both model and parametric uncertainties may be treated using this approach, in practice it is not clear that separating MST consequence uncertainties into parametric and model uncertainties will make a significant difference as far as Monte Carlo analysis is concerned. The MHTGR and PBMR NGNP radionuclide retention uncertainty analyses used parameters to model the uncertainty in processes for which physical models were unavailable. For each item a combination of model and parametric uncertainty was used, or only parametric uncertainty was used, in which case the model was comprised of a simple mass balance. Examples included:

a. The fraction of fuel particles that were failed at the beginning of the event and during the event in each spatial point in the reactor core

b. The fraction of radionuclides released from intact and failed fuel particles in each spatial point in the reactor core

c. The fraction of core at time and temperature in each spatial point in the reactor core

d. The distribution of released radionuclides into elemental, aerosol and particulates in each volume of the reactor core, the helium pressure boundary, and in the reactor building

e. The location and size of the leak/ breach in the helium pressure boundary

f. The location and size of the leak paths in the reactor building and the response of blowout panels and dampers

Based on this experience it was found that the most important consideration is that the key sources of uncertainty have been identified and accounted for in the quantification of uncertainty or in the subsequent sensitivity analyses. Because some sources of uncertainty could alternatively be addressed with parameters, models, or parametric models, how the sources are sorted into these categories was not found to be as critical.

As the MST consequence evaluations proceed in parallel with model development from the design effort and with design data need completion from the technology development program, the NGNP Project will evaluate the need for more than one detailed model for various phenomena. The relative contributions of parametric and model uncertainty to the overall uncertainty in MSTs and dose consequences will be considered taking into account the information provided above. Regardless, as discussed in the response to RAI MST-88 the uncertainty analyses will, of necessity for the Monte Carlo evaluation, utilize a simplified overall model in comparison to the detailed models for each of the phenomena, e.g., thermal, reactivity, chemical, mass transport, dose, etc..

**REQUEST REGARDING FOLLOWUP ITEM FQ/MST-29:**

During its review of the Fuel Qualification and Mechanistic Source Terms White Papers, the NRC staff identified several follow up items that it intends to pursue as the project proceeds. These items have been addressed in a series of meetings between the NRC staff and NGNP personnel. One of the items identified by the staff, referred to as Follow up Item FQ/MST-29, is the following:

> Long Term Modeling of Radionuclide Transport within the Core and the Reactor Coolant System
>
> *A great deal of discussion is provided in the white papers on experiments and modeling radionuclide release from the fuel. However, much less discussion is given to source term model development and verification beyond the fuel such as transport in the reactor system and behavior following release from the reactor system. Indeed, a major challenge in the accident analysis of modular HTGRs is the modeling of radionuclide transport within the core and the reactor coolant system over many years of normal plant operation before initiation of an accident or transient. This is an item for follow up.*

In the July 24, 2012 NGNP/NRC meeting on fuel qualification and mechanistic source terms, the NGNP staff documented the following discussion of this item and committed to the following action:

> NRC stated that it would like to receive additional information regarding which radionuclides are expected to be the predominate contributors to offsite doses for various HTGR accident sequences. NRC would also like information on the relative inventories of these isotopes that are expected to be present in the core, in the helium coolant or plated out on helium pressure boundary surfaces, released to the reactor building, and released to the environment under various accident sequences. NGNP stated that this information is available for the GA MHTGR design in the PSID and agreed to provide a summary of the requested information for the staff.

**RESPONSE:**

<u>Radionuclides Contributing to Onsite and Offsite Dose</u>

As described in the Mechanistic Source Terms White Paper (MST WP 2010), the design of the NGNP modular HTGR is still being defined, and design-specific source terms have not yet been calculated. Consequently, the above request for information cannot rigorously addressed for the NGNP at this writing. However, a good indication of the expected dominant dose contributors and the radionuclide distributions throughout the plant can be inferred from the extensive plant design and safety analyses that were completed in the late 1980s for the 350 MW(t) MHTGR that were documented in a Preliminary Safety Information Document submitted to the NRC (PSID 1992).

The key radionuclides that dominated the radiological doses (offsite doses from accidents and from normal operation, and worker doses) predicted for the MHTGR are listed in Table 1. This list was developed from information that was reported throughout (PSID 1992), including in Section 4.2.5.2.2, Fuel Performance; Section 9.1.2, Helium Purification System; Section 11, Operational Radionuclide Control; Section 12, Occupational Radiation Protection; and Section 15, Safety Analysis. The PSID also includes a number of appendices providing responses to a

large number of NRC comments/questions; these responses also include information about dominant dose contributors, especially the responses to comments R 4-7, R 4-9, R 11-9, and R 15-13.

A broad spectrum of radionuclide control requirements were imposed upon the MHTGR design (PSID 1992, Section 4.1.3). The bounding requirement was meeting the EPA Protective Action Guides (PAGs) at a 425-m Exclusion Area Boundary (EAB); specifically, the thyroid PAG (5 rem) was more constraining than the whole-body PAG (1 rem). The second most constraining, top level radionuclide control requirement was to limit the occupational exposure to ≤10% of 10CFR20. Typically, occupational exposures result primarily from O&M activities, especially in-service inspection (ISI), during normal plant operation.

The most important off-site dose contributor is I-131 (and to a lesser extent other radioiodine isotopes) because of the PAG thyroid dose limit (Table 1). The requirement to meet the PAGs during depressurization events without forced cooling is used to set limits on the accumulation of I-131 during normal operation. The iodine release limits are the most constraining on the fuel design for both normal operation and for a broad spectrum of postulated accidents (PSID 1992, Section 4.2, Section 15). The limits on noble gas release, which are less constraining on the fuel design than are the iodine release limits, are derived from whole-body PAG dose limits. For short-term events, such as rapid depressizurations, 2.8-hr Kr-88 is the dominant noble-gas dose contributor for the early release, and 5.3-d Xe-133 is inconsequential by comparison. For long-term events, such as slow depressurizations, their importance for early release is reversed (because of the short half life of Kr-88). Likewise, the contributions of Cs-137 and Ag-110m to offsite doses are small compared to that from I-131 (PSID 1992, Section 15, Response to Comment R 4-7).

Ag-110m is not a significant contributor to offsite doses during normal operation or accidents. Its importance is as a contributor to occupational doses during plant maintenance and ISI as a result of its multiple hard gamma emissions (PSID 1992, Section 4.2.5.2.2.2.5). The production of Ar-41 in the cooling air passing through the Reactor Cavity Cooling System (RCCS) (for those designs that feature RCCS with air cooling) is a significant contributor to the normal offsite dose from gaseous effluents (PSID 1992, Section 11.1.3.1). H-3 is the dominant contributor to the normal offsite dose from liquid effluents (PSID 1992, Section 11.1.4).

The above discussion of key radionuclides is specific to the steam-cycle MHTGR. Similar circumstances are expected for the NGNP modular HTGR (MST WP 2010).

Normal Operation Radionuclide Distributions – Core, Coolant, and Plateout

As stated above, the design of the NGNP modular HTGR is still being defined, so a design-specific, overall plant mass balance for radionuclides is not available at this writing. However, the radionuclide plant mass balance for the 350 MW(t) MHTGR presented in (PSID 1992) is a good general indicator of the mass balance that will ultimately be calculated for NGNP modular HTGR.

As described in (PSID 1992, Section 11.1) and in (MST WP 2010), standard design practice in the U.S. HTGR program has been to define a two-tier set of radionuclide design criteria, referred to as "Maximum Expected" and "Design" criteria. The fuel and core are to be designed such that there is at least a 50% probability that the fission product release will be less than the "Maximum Expected" criteria and at least a 95% probability that the release will be less than the "Design" criteria.

The "Maximum Expected" radionuclide design criteria for the 350 MW(t) MHTGR for the radionuclides that are the major contributors to offsite doses are summarized in Table 2. (The "Maximum Expected" core inventories shown in Table 2 are not in the PSID. The core inventories given in Table 2 were taken from the same analysis that was used to generate "Design" inventories given in PSID Section 11.1.) The corresponding "Design" criteria are included in (PSID 1992, Section 11.1). Table 2 presents the relative distribution of each radionuclide among the core, the coolant, and the primary coolant surfaces for the MHTGR. Information is also provided on the distribution of the core inventory among intact fuel particles, fuel particles with various defects, the fuel compact matrix, and the fuel element structural graphite.

It is anticipated that the relative radionuclide distributions in the NGNP modular HTGR will be quite similar to that presented in Table 2 for the MHTGR. However, there will be some differences between the two designs that will change the absolute Ci inventories. The inventories scale proportionally with total power. The core inventories scale as the product of the fuel residence time and the capacity factor (the expected higher capacity factor in the NGNP would more than compensate for the shorter expected fuel residence time). The longer expected plant life time for the NGNP modular HTGR (60 yr vs. 40 yr) only affects the plateout inventories of the long-lived radionuclides (e.g., 30-yr Cs137, 28-yr Sr-90, etc.).

There could be further differences depending on design decisions made by the NGNP modular HTGR designer. However, the information presented here is generally expected to be representative of results to be obtained for the NGNP modular HTGR.

Radionuclide Releases Under Accident Conditions

The distribution of radionuclides in the core, in the coolant, and on primary coolant surfaces defines the initial conditions for analyses of radionuclide release under accident conditions. Radionuclide releases from the MHTGR under various limiting Design Basis Accident conditions are described in (PSID 1992, Section 15.13).

Three accident sequences were identified in the PSID as being limiting with respect to offsite doses. These accidents are referred to and described in the PSID as follows:

*SRDC-6, Depressurized Conduction Cooldown with Moderate Moisture Ingress:*

A steam generator tube leak results in moisture ingress into the reactor system. Moisture ingress is followed by depressurization of the helium coolant and loss of forced cooling. Reactor trip on outer control rods is provided by high core power-to-flow ratio, as a result of the rapid increase in reactivity due to moisture. Also, the reserve shutdown control material is inserted and the main loop is shut down in response to high primary coolant pressure. The reactor cavity coolant system is effective in removing the decay heat.

The radiological impact of SRDC-6 is attributed to the release of circulating activity, steam-induced vaporization (washoff) and recirculation of plated-out activity, release of a small fraction of the fuel activity due to the core temperature transient, release of fission products contained in the matrix and structural graphite that becomes oxidized, and release due to the hydrolysis of failed fuel. The recirculation of plateout and the hydrolysis of failed fuel before the depressurization are the major contributors to iodine and noble gas release. Delayed release from the fuel during core heatup is also a major contributor to radionuclide release. Dose to a

receptor at the EAB is calculated considering, mechanistically, the phenomena of plateout and settling in the Reactor Building, radioactive decay, and atmospheric dispersion.

*SRDC-10, Depressurized Conduction Cooldown with Moderate Primary Coolant Leak:*

A leak of 81.9 sq cm (12.7 sq in.) occurs at the top of the steam generator vessel, corresponding to a failure of the pressure relief train. The leak depressurizes the Reactor System in minutes. At 20 seconds, on a low-pressure signal, the reactor is tripped using the outer control rods. The heat transport system is tripped soon afterwards on another even lower pressure signal and quickly coasts down to a no-forced-circulation condition. No forced circulation cooling is available to the reactor, resulting in a gradual heatup and cooldown of the core and its surroundings. The reactor cavity cooling system successfully removes heat by radiation and conduction.

Radiological release during a depressurized conduction cooldown with moderate primary coolant leak is attributed to the circulating activity, the liftoff of a portion of the plated-out activity, and the release of some small fraction of fuel activity. During the initial depressurization, most of the circulating and liftoff activities are released. Release from the fuel occurs during the gradual heatup of the core and is the largest contributor to the offsite dose for this event. Dose to a receptor at the EAB is calculated considering mechanistically the phenomena of plateout and settling in the Reactor Building, radioactive decay, and atmospheric dispersion.

*SRDC-11, Depressurized Conduction Cooldown with Small Primary Coolant Leak:*

A leak of 0.32 sq cm (0.05 sq in.), which corresponds to the size resulting in the greatest release, occurs at a penetration at the top of the reactor vessel. The leak depressurizes the Reactor System in 24 hours. At 35 minutes, on low-pressure signal, the reactor is tripped using the outer control rods. After reactor trip forced circulation cooling to the reactor is no longer available, resulting in a gradual heatup of the core and its surroundings. Since natural circulation within the core is ineffective during depressurization, the core thermal transient of SRDC-11 is identical to that of SRDC-10. The reactor cavity cooling system successfully removes heat by radiation and conduction.

Radiological release during a depressurized conduction cooldown with a small primary coolant leak is attributed to the circulating activity and the delayed release of some small fraction of fuel activity. During the slow depressurization, both sources of fission products contribute to the release, with the delayed release being the dominant contributor to offsite dose. Dose to a receptor at the EAB is calculated considering mechanistically the phenomena of plateout and settling in the Reactor Building, radioactive decay, and atmospheric dispersion.

Table 3 is a summary of information available from (PSID 1992, Section 15.13) on the release of dominant radionuclides from the MHTGR vessel system to the reactor building for these three limiting accident sequences. Releases are presented for the short term release from the depressurization, for the longer term delayed release from heatup of the reactor core, and for the total duration of the accident sequence. This information indicates that, for each accident sequence, the longer term delayed release during core heatup is the larger contributor to the most limiting offsite doses from iodine.

Radionuclide holdup and depletion in the reactor building is addressed in (PSID 1992, R 15-1 and R 15-12). As noted in the PSID, the MHTGR meets the regulatory requirements for offsite dose even at the EAB for all Licensing Basis Events (LBEs) with at least 50% confidence

assuming no reactor building attenuation. However, accounting for attenuation in the reactor building allows the 5 rem thyroid PAG to be met at the EAB.

Table 4 presents a summary of information available from the PSID on mean thyroid dose at the EAB for these three limiting accident sequences. Information is presented for analyses with and without taking credit for attenuation in the reactor building. This information indicates that the iodine attenuation factor for the reactor building varies with the accident sequence, ranging from a factor of 4 to a factor of 17 for these limiting accident sequences.

Table 4 also shows that the thyroid dose at the EAB for these accident sequences is substantially below the 300 rem limit of 10CFR100.11, both with and without taking into account radionuclide attenuation in the reactor building.

## REFERENCES

(MST WP 2010) "Mechanistic Source Terms White Paper," INL/EXT-10-17997, Rev. 0, Idaho National Laboratory, July 2010.

(PSID 1992) "Preliminary Safety Information Document for the Standard MHTGR," HTGR-86024, Rev. 13, Stone & Webster Engineering Corp., September 1992.

**Table 1. Key Radionuclides in HTGRs**

| Nuclide | Half Life | Nuclide Class* | Primary Production | Dominant Source | Primary Impact |
|---|---|---|---|---|---|
| **I-131** | 8 day | Halogen | (U,Pu) fission | HM contamination<br>Exposed kernels | Offsite dose during accidents (dominant)<br>O&M worker dose |
| **Ag-110m** | 250 day | Noble metal | Pu fission → $^{109}Ag$<br>$^{109}Ag(n,\gamma)^{110m}Ag$ | Diffusive release from intact TRISO particle | O&M worker dose |
| **Cs-137** | 30 yr | Alkali metal | (U,Pu) fission | Exposed kernels | O&M worker dose, offsite dose during accidents |
| **Cs-134** | 2.1 yr | Alkali metal | (U,Pu) fission | Exposed kernels | O&M worker dose, offsite dose during accidents |
| **Sr-90** | 28 yr | Alkaline earth | (U,Pu) fission | Exposed kernels | Offsite bone dose during accidents |
| **Kr & Xe** | -- | Noble gas | (U,Pu) fission | Heavy Metal contamination<br>Exposed kernels | Offsite dose during accidents (minor)<br>Normal operation gaseous effluent offsite dose |
| **H-3** | 12.3 yr | Tritium | Multiple sources; primary:<br>$^{3}He(n,p)^{3}H$ | Neutron activation of $^{3}He$ in primary coolant: | Normal operation liquid effluent offsite dose |
| **Ar-41** | 1.8 hr | Noble gas | Air activation in air-cooled versions of the RCCS | Air-cooled versions of the RCCS | Normal operation gaseous effluent offsite dose |

*As defined in NGNP MST White Paper [MST WP 2010].

**Table 2. Distribution of Radionuclides in 350 MW (t) MHTGR for Major Contributors to Offsite Dose During Accidents ("Maximum Expected" Inventories)**

| Radionuclide | "Maximum Expected" Inventory (Ci) | | | | |
|---|---|---|---|---|---|
| | **Kr-88** | **Xe-133** | **I-131** | **Sr-90** | **Cs-137** |
| | | | | | |
| Intact Particles | $9.9 \times 10^{6}$ | $2.0 \times 10^{7}$ | $9.3 \times 10^{6}$ | $7.4 \times 10^{5}$ | $8.6 \times 10^{5}$ |
| Defective SiC | $5.0 \times 10^{2}$ | $1.0 \times 10^{3}$ | $4.7 \times 10^{2}$ | $3.7 \times 10^{1}$ | $4.3 \times 10^{1}$ |
| Exposed Kernels | $4.9 \times 10^{2}$ | $9.7 \times 10^{2}$ | $4.5 \times 10^{2}$ | $3.6 \times 10^{1}$ | $2.2 \times 10^{1}$ |
| Fuel Compact Matrix | $9.4 \times 10^{1}$ | $1.8 \times 10^{2}$ | $8.2 \times 10^{1}$ | 7.3 | 20 |
| Graphite | - | - | - | 1.2 | 10 |
| **Equilibrium Total Core** | $9.9 \times 10^{6}$ | $2.0 \times 10^{7}$ | $9.3 \times 10^{6}$ | $7.4 \times 10^{5}$ | $8.6 \times 10^{5}$ |
| | | | | | |
| **Primary Circuit** | | | | | |
| Plateout (40-yr) | - | - | 20 | 0.34 | 70 |
| Circulating | 5.2 | 2.3 | 0.018 | $7.3 \times 10^{-10}$ | $2.0 \times 10^{-6}$ |

**Table 3. Expected Radionuclide Releases (Ci) to Reactor Building for Limiting MHTGR Design Basis Accidents**

| Radionuclide | Kr-88 | I-131 |
|---|---|---|
| **Accident** | | |
| Short Term Release from Depressurization | | |
| SRDC-6 | 20 | 18 |
| SRDC-10 | <1 | <1 |
| SRDC-11 | <1 | <1 |
| Delayed Release from Fuel Heatup | | |
| SRDC-6 | <1 | 32 |
| SRDC-10 | 6 | 26 |
| SRDC-11 | 10 | 95 |
| Total Release | | |
| SRDC-6 | 20 | 50 |
| SRDC-10 | 6 | 26 |
| SRDC-11 | 10 | 95 |

**Table 4. Mean Thyroid Dose at the EAB for Limiting MHTGR Design Basis Accidents With and Without Reactor Building Attenuation (rem)**

| | With Attenuation | Without Attenuation |
|---|---|---|
| **Accident** | | |
| SRDC-6 | 1.1 | 5 |
| SRDC-10 | 0.041 | 0.7 |
| SRDC-11 | 0.73 | 3 |