Release Fractions in Non-LOCA Accidents in Draft Regulatory Guide 1.183 DG-1199

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Cesium Release Fraction

In this note, an argument is made that the non-loss of coolant accident (LOCA) fractions of fission product inventory found within the gap is too high for alkali metals as currently specified within Regulatory Positon 3.2 of Draft Regulatory Guide 1.183 rev.1 (DG-1199). This assertion extends to the enthalpy-dependent transient fission product release component used in reactivity initiated accidents.

Regulatory Position 3.2 of Draft Regulatory Guide 1.183 rev. 1, which is presented below, details the release fractions of fission product inventory for postulated accident scenarios including both LOCAs and non-LOCA accidents. Relevant non-LOCA accidents include fuel handling accidents, boiling water reactor (BWR) rod drop accidents, pressurized water reactor (PWR) rod ejection accidents, BWR/PWR main steam line breaks, PWR steam generator tube ruptures and PWR locked rotor accidents. In the regulatory position below, Table 3 refers to Table 1 in this note and Table 4 refers to Table 2.

> "For non-LOCA DBAs other than reactivity-initiated accidents (RIAs), where only the cladding is postulated to be breached, Table 3 gives the fractions of the core inventory for the various radionuclides assumed to be in the gap for a fuel rod. The release fractions from Table 3 are used in conjunction with the calculated fission product inventory calculated with the maximum core radial peaking factor. [omitted]
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> For RIAs, such as the BWR control rod drop accident and PWR control rod ejection accident, the total fraction of fission products available for release equals the steady-state fission product gap inventory in Table 3 plus the transient fission product release resulting from the rapid power excursion. Table 4 lists the combined fission product inventory, by radionuclide groups, available for release for a fuel rod during an RIA. The transient fission product release component is presented as a function of increase in radial average fuel enthalpy (ΔH, calories per gram (cal/g)). This component of the overall fission product inventory may be calculated separately for each axial node which experiences the RIA power pulse and then combined to yield the total transient fission product release for a particular fuel rod. The sum total of combined fission product inventories from each fuel rod predicted to experience cladding failure (all failure modes) should be used in the dose assessment.
>
> [omitted paragraph]

> The non-LOCA fission product gap inventories listed in Table 3 and RIA combined release fractions listed in Table 4 do not include the additional contribution associated with fuel melting. The event-specific appendices provide guidance for adjusting these gap inventories for fuel rods that are predicted to experience limited fuel centerline melting."

For the postulated non-LOCA accident the fraction of fission product inventory in the gap that is assumed to be released is specified for I-131, I-132, Kr-85, other noble gases, other halogens and alkali metals. Alkali metals includes cesium. The values in the regulatory guide for the inventory in the gap can be seen in Table 1.

Table 1. Non-LOCA Fraction of Fission Product Inventory in Gap for DG-1199

Group	Fraction
I-131	0.08
I-132	0.09
Kr-85	0.38
Other Noble Gases	0.084
Other Halogens	0.05
Alkali Metals	0.50

For reactivity insertion accidents a further enthalpy dependent factor is applied to the inventory, assuming a finite amount of fuel melting from the addition of reactivity. For these accidents the combined release fractions can be seen in Table 2.

Table 2. Fraction of Fission Product Inventory Available for Release from Reactivity Initiated Accidents for DG-1199

Group	Combined Release Fraction*
I-131	[(0.08) + (0.00073 * ΔH)]
I-132	[(0.09) + (0.00073 * ΔH)]
Kr-85	[(0.38) + (0.0022 * ΔH)]
Other Noble Gases	[(0.084) + (0.00073 * ΔH)]
Other Halogens	[(0.05) + (0.00073 * ΔH)]
Alkali Metals	[(0.50) + (0.0031 * ΔH)]

$* \Delta H = increase\ in\ radial\ average\ fuel\ enthalpy, cal/g$

When published as Regulatory Guide 1.183 (DG-1081) in July 2000 the values for non-LOCA fraction of fission product inventory in the gap were different. The original values can be seen in Table 3. When comparing the updated table to the original one, it can be seen that the releases of Kr-85 and alkali metals increased significantly. For Kr-85, the increase is from a release fraction of 0.10 to a release fraction of 0.38. For alkali metals, which includes Cs, the increase is from 0.12 to 0.50.

Table 3. Non-LOCA Fraction of Fission Product Inventory in Gap for DG-1081

Group	Fraction
I-131	0.08
Kr-85	0.10
Other Noble Gases	0.05
Other Halogens	0.05
Alkali Metals	0.12

The methodology and calculation of the updated values is described in "Update of Gap Release Fractions for Non-LOCA Events Utilizing the Revised ANS 5.4 Standard," also listed as PNNL-18212 Rev. 1. The information contained in these tables was based on calculations performed using the ANS5.4-2011 standard equations implemented into the fuel performance code FRAPCON3.3 to determine release-to-birth ratios of key isotopes. The release to birth ratio is defined as the ratio of total atoms released to gap over the total number of atoms produced. This ratio is directly used to determine the fission product inventory in the gap. (PNNL-18212 Rev. 1)

The document ANS5.4-2011 is an update of ANS5.4-1982. Since the publication of ANS5.4-1982, significant research at Halden was performed into better understanding the behavior of short-lived isotopes a gap release accident. (White and Turnbull, 1998) (PNNL-18212 Rev. 1) Additionally, there was significant work on extending the modeling of fission gas release to high burnup fuel.

The higher values of Kr-85 release in DG-1199 can be attributed to higher burnups that needed to be considered in order to create a bounding case for the regulatory guide. It can be seen that at high burnup the predicted release fraction for Kr-85 increases to a peak near a rod-average burnup 55 MWd/kgHM. This can be seen in Figure 1.

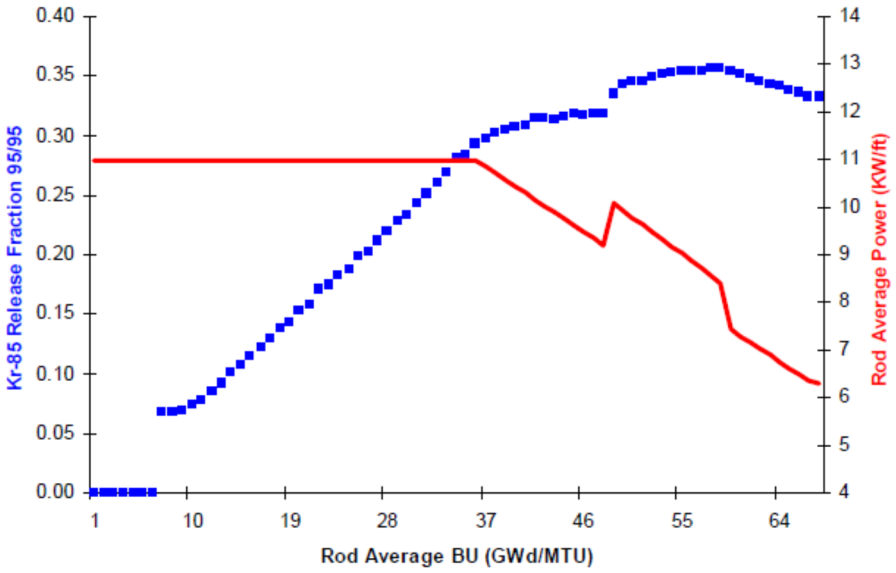

Figure 1. Predicted Release Fraction for Kr-85 (Long-Lived) Isotope as a Function of Burnup Illustrating Peak Release Occurs Near EOL (Figure 2.5 in PNNL-18212 rev. 1)

Per the technical basis document, this Kr-85 release was calculated using the fuel performance code FRAPCON 3.3 with associated uncertainties. Alkali metals release-to-birth ratios, unlike those for noble gases are not calculated directly by FRAPCON. Instead, scaled releases are estimated using Equation 1 and Equation 2. (PNNL-18212 Rev. 1)

$$Cs^{134}(Gap\ Release) = Kr^{85}(Gap\ Release) \times (2)^{0.5} + Uncertainty \qquad \text{(Eqn. 1)}$$

$$Cs^{137}(Gap\ Release) = Kr^{85}(Gap\ Release) \times (2)^{0.5} + Uncertainty \qquad \text{(Eqn. 2)}$$

The factor of $\sqrt{2}$ is to account for cesium's assumed diffusion coefficient, which is taken to be 2.0 times higher than noble gasses. Release fraction is approximately proportional to the square of the diffusion coefficient. This treatment of cesium release in relation to Kr-85 release is also used in to express the enthalpy dependent portion of release for non-LOCA reactivity insertion accidents. See Equations 3 and 4. (PNNL-18212 Rev. 1)

$$F(Cs) = F(Kr^{85}) \times (2)^{0.5} \times \Delta H \qquad \text{(Eqn. 3)}$$

$$F(Cs) = 0.0022 \times (2)^{0.5} \times \Delta H = 0.0031 \times \Delta H \qquad \text{(Eqn. 4)}$$

This treatment is a holdover from ANS5.4-1982. The rationale for this treatment is detailed in Section 2.2.4 of PNNL-18212 Rev. 1, which is contained below.

> "As recommended by the 2011 ANS 5.4 standard the cesium diffusion should be assumed to be a factor of 2 higher than for the noble gases (xenon and krypton), which has been carried over from the 1982 ANS 5.4 standard because the current ANS 5.4 Working Group was not aware of any new quantitative cesium release data under normal LWR operating conditions."

Tying alkali metal (and Cs) release directly to Kr-85 release leads the release of cesium to be extremely high relative to other known measurements of Cs release behavior from experiments. Moving from DG-1081 to DG-1199, the alkali metal release fraction increased by nearly a factor of four, from 0.12 to 0.50. It is believed that an increase this large without a justification that is based on cesium-specific data does not reflect current understanding and could be excessively conservative.

Additionally, while ANS5.4-2011 indicates that there was no new quantitative information on cesium release under normal LWR operating conditions, no mention was made of numerous experiments on fission product release. (PNNL-18212 Rev. 1) Of particular note are the Phebus and VERCORS experiments which had been performed and used as the technical basis for severe accident analysis codes such as MELCOR (Gauntt, 2010).

One of the key insights from these experiments is that the release rate of cesium relative to the rate of release of noble gases changes as a function of fuel burnup. This behavior was

summarized by Powers and Bottomly (2010). At lower burnups cesium behavior and release tracks noble gas release very closely. However, as the burnup of the fuel increases; the ability of cesium that is produced within the fuel to be released from the gap is inhibited by two different processes. First, there is holdup on the grain boundaries of the fuel matrix by interactions with hyperstoichiometric uranium dioxide, UO_{2+x}, and molybdenum oxide, MoO_2, to form cesium-uranium oxide compounds and cesium molybdate. Both the formation of hyperstoichiometric uranium dioxide and molybdenum oxide increase with burnup. Second, if cesium migrates from grain boundaries to the gap region there is the potential to interact with a layer of zirconium dioxide on the internal layer of the cladding, which forms at higher burnups. This layer of ZrO_2 interacts with the migrating cesium to form cesium zirconates, further inhibiting the release of cesium. (Powers and Bottomly, 2010)

Given that both of these phenomena limit the volatility of cesium at higher burnups, an argument for directly tracking cesium release with noble gas release cannot be made. Noble gas release clearly increases at higher burnups; however, that is not necessarily the case with cesium.

Additional analyses by Powers (2013) concluded that based on the diffusion characteristic of key cesium species through a helium gas in relation to the diffusion of the noble gas xenon, the total diffusion rate of cesium species would be less than that of xenon.

$$D_{Xe,He} \geq D_{Cs,He} \geq D_{CsMo_4,He} \qquad \text{(Eqn. 5)}$$

It is recommended that in lieu of scaling the cesium release with Kr-85 a physics-based or empirical data-based chemical transport model for cesium release be used. Should the cesium release be calculated based on the release of a noble gas, it is recommended that the cesium release be based on that of xenon and not krypton. Xenon is included in the "other noble gases" listed in Tables 1, 2 and 3.

It is also recommended that the relation between Kr-85 and cesium in ANS5.4-2011 should be altered to better reflect experimental data.

Recommended Update of Non-LOCA Fragmentation-Induced Fission Gas Release Augmentation Factors

New regulatory guidance within Clifford (2015) indicates that a revision of the fragmentation-induced fission gas release fractions found within Table 4 of DG-1199 requires an update. (See Table 2 of this note.) This update is only pertinent to reactivity insertion accidents, and hence only applies to Table 4 of the document. Within Clifford (2015) new experimental data is presented and proposed changes to the fragmentation-induced factors are made. It is recommended that these changes be implemented within DG-1199. Specifically, it is recommended that the "Kr-85" and "Other Noble Gas" entries of Table 4 in DG-1199 be

adjusted as follows, with two separate values for low/medium and high burnups. It is also recommended that the "Alkali Metals" entry be updated using the previously discussed scaling factor of $2^{0.5}$. However, this is recommended to maintain consistency with the treatment of "Alkali Metals" elsewhere in the regulatory guide. It is maintained, per the earlier discussion, that scaling according to this factor is not adequately justified.

Peak Pellet Burnup ≥ 50 GWD/MTU:

Kr-85:	((0.38) + (0.0022 * ΔH)) ➔ ((0.38) + (0.0026 * ΔH - 0.05))
Other Noble Gases:	((0.084) + (0.00073 * ΔH)) ➔ ((0.084) + (0.0026 * ΔH - 0.05))
Alkali Metals:	((0.50) + (0.0031 * ΔH)) ➔ ((0.084) + (0.0037 * ΔH - 0.07))

Peak Pellet Burnup < 50 GWD/MTU:

Kr-85:	((0.38) + (0.0022 * ΔH)) ➔ ((0.38) + (0.0026 * ΔH - 0.13))
Other Noble Gases:	((0.084) + (0.00073 * ΔH)) ➔ ((0.084) + (0.0026 * ΔH - 0.13))
Alkali Metals:	((0.50) + (0.0031 * ΔH)) ➔ ((0.50) + (0.0037 * ΔH - 0.18))

These two separate correlations follow the experimental data closely, as can be seen in Figure 2, where both are applied against multiple data sets for different reactor and fuel types.

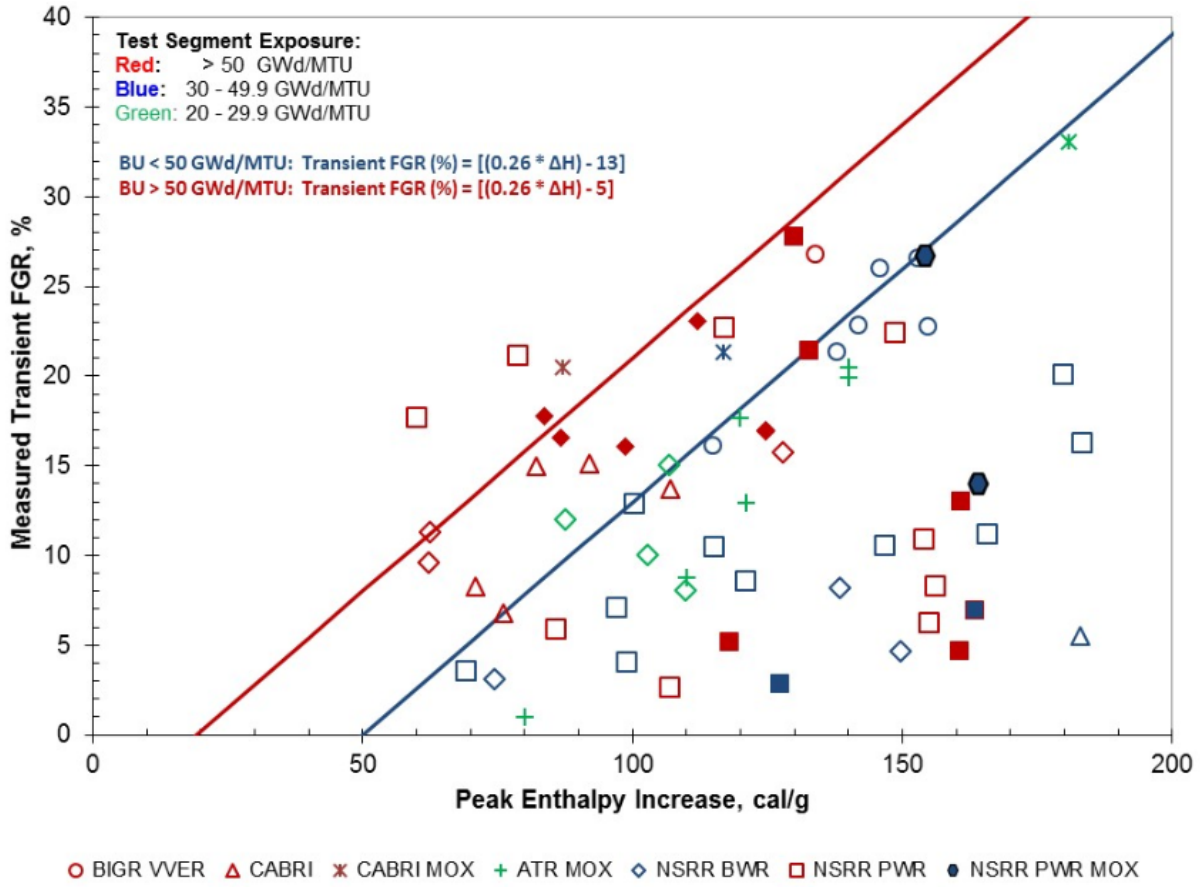

Figure 2: Burnup-Dependent Transient FGR Correlations. Open symbols indicate non-failed, solid symbols indicate cladding failure. (Clifford, 2015)

References:

American Nuclear Society, "ANSI/ANS-5.4-2011: Method for Calculating the Fractional Release of Volatile Fission Products from Oxide Fuel," Issued as an ANS Standard, 2011.

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