Response of Nuclear Materials in Storage and Transport to Malevolent Environments

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Abstract

Work related to the assessment of radiological health consequences resulting from a sabotage attack on nuclear fuel storage or transport casks has been on-going since the late 1970's. While the level of effort in this area has been uneven over these three decades due to policy priorities, funding levels, and programmatic priorities of the countries funding this type of work, substantial progress has been made. From phenomenology of in-cask transport processes to development of aerosol production in high energy attack environments, the analytical and experimental work performed provides substantial justification to consequence assessments that heretofore have had to rely on conservative assumptions in lieu of empirical data.

One constant since the late 1990's in addressing this problem has been an international working group whose primary focus has been to develop source term data from experimental simulations of sabotage-types of attacks. This working group, titled; the International Working Group for Sabotage Concerns of Transport and Storage Casks (WGSTSC) is comprised of experts mainly from the U.S., France, and Germany. Technical support has also been provided, on an intermittent basis, from the U.K. and Japan. The WGSTSC has pooled resources and expertise to design and conduct experiments that produce the data needed to perform radiological consequence assessments. In addition to the experimental program, this group has also coupled modern analytic techniques with experimental results to understand the effects of high energy density devices on nuclear materials.

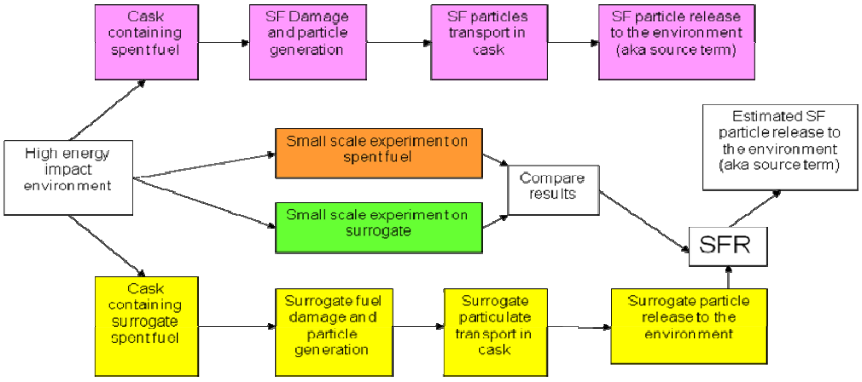
This paper will review the paradigm that has guided the WGSTSC effort and show how the results from the experimental programs of the past 3 decades has brought us to the current level of understanding of the potential consequences from a malevolent attack on nuclear transport and storage facilities. In addition, the paper will provide an update on the status of the work of the WGSTSC and describe what additional experimental and analytical efforts would be most productive in further narrowing of uncertainty in consequence prediction.

Introduction

Estimation of the consequences of a sabotage attack on nuclear materials in transport requires an understanding of the amount and physical/chemical form of radioactive materials released as a result of the attack. The amounts released are a function of the characteristics of the material and the mode of attack. In addition, the characteristics of the package, as they are affected by the attack mode, provide limitations on the amount of the active materials released through the damaged areas of the packaging[1].

Were the materials involved non-radioactive, it would be a relatively simple matter to perform experiments to obtain the needed information, but experimentation with real spent fuel in association with high explosives presents a combined hazard for which there is essentially no large scale test facility in which to test. As a result, the information must be developed in a manner as indicated in Figure 1. Instead of a direct experiment as shown in the upper sequence, surrogate material (usually DUO_2) in real casks subjected to real sabotage modalities is used for basic aerosol generation data and for information on how the aerosols escape to the environment from a damaged cask (lower sequence). Then experiments must be done to relate the behavior of real spent fuel exposed to similar high energy environments to that for DUO_2 in the same environment (middle sequence). The latter two paths have been followed in pursuing this research effort since the late 1970's.

Figure 1. Schematic of Source Term Estimate Process



Overview of Prior Analyses and Research Efforts

Between 1975 and 1977 the US Nuclear Regulatory Commission (NRC) sponsored work at Sandia National Laboratories (SNL) to evaluate the risk to the public of shipping radioactive materials which culminated in the publication of NUREG-0170, "Final Environmental Statement on the Transportation of Radioactive Materials by Air and Other Modes". While no specific consideration of shipment sabotage was included in the report, potential risks relating to consequences of sabotage were raised for a future study more specific to the issue. In addition, it was noted that the NUREG-0170 analyses did not explicitly treat very high population density areas (like city centers). To better understand these issues,

[1] Within a package the aerosol produced is the aerosolized fraction (AF) of the affected mass. The fraction of that in the respirable size range is the respirable fraction, RF. A fraction of the aerosol within the package may be released to the environment leading to the respirable released fraction, RRF and the aerosol released fraction, ARF. Frequently the mass or activity released is given as a fraction of the entire content of the cask (RRFc and ARFc) a fact that requires the reader to be aware of the context of the numbers being discussed.

the NRC went forward with internal studies and tasked SNL to look at transport safety issues in urban areas which lead to the "Urban Study" discussed below.

In 1977, Hodge and Campbell (of the NRC) published a study sponsored by the NRC Division of Safeguards, entitled *Calculations of Radiological Consequences from Sabotage of Shipping Casks for Spent Fuel and High-Level Waste* (Hodge, 1977). Without delineation of threat and mode of attack, the authors judged "massive rupture" of a spent fuel cask to be "incredible" and postulated a small penetration that could result in the release of a maximum of 1% of the total fuel solids and 100% of the gases (e.g., krypton-85). The effect of selectively raising the release fraction of specific volatile isotopes (radio-cesium and radio-tellurium) to 1.0 was also examined. The maximum release (in which 100% of volatiles were assumed to be released) yielded an estimate of 2 early fatalities and 40 to 260 latent cancer fatalities in an area with a population density of 100 persons/mi^2 (typical of US average population density). In an alternative scenario, the release fractions of all isotopes were set to 1% except for noble gases, which remained at 100%. For this scenario, an estimate of zero early fatalities and 40 latent cancer fatalities was obtained. The radionuclide inventory used for the consequence estimates was for short-cooled (150 days out of reactor) spent fuel as might have been shipped in a fuel cycle with reprocessing.

The "Urban Study" had a preliminary version (DuCharme, 1978) and a final report (Finley, 1980) that considered the potential consequences of a successful sabotage act in a major urban area. An analysis of potential consequences of sabotage contained in the preliminary version of the Urban Study was based on a very conservative set of analytical assumptions utilizing upper limits for the various parameters involved in the consequence calculation. The results presented predicted tens of early fatalities and hundreds to thousands of latent cancer fatalities (LCF) from an optimally effective attack scenario in a location with a very high population density such as the Borough of Manhattan in New York City. In response to the very high potential doses in the preliminary version of the Urban Study, the NRC instituted a rulemaking to require greatly enhanced security for fuel shipments passing through urban areas.

The final version of the Urban Study recognized the conservative nature of the assumed source term in the preliminary version and repeated the impact estimates with a somewhat more realistic and significantly smaller respirable release (RRFc $\approx 10^{-3}$) that resulted in less than 1 early fatality, and 10 to 100 LCF.

In light of the uncertainty in release fractions used in the Urban Studies and in the Hodge and Campbell report, in the late 1970s and early 1980s, NRC sponsored experimental research at Battelle Columbus Laboratories (BCL) to get a better estimate of the effects of HEDD environments on spent fuel and how that might relate to package release fractions. The Department of Energy (DOE), sponsored similar work at SNL to better define the release fraction as well in the expectation that a demonstrably smaller source term would be an argument to reduce the stringency of the new NRC security requirements.

BCL Program: - A total of 8 tests were performed in which a small HEDD was fired to penetrate a mock-up cask wall and impact segments of *actual* pressurized water reactor (PWR) spent fuel rods (Schmidt, 1982). As part of the test apparatus checkout, 6 tests using depleted uranium pellets as a surrogate for spent fuel were carried out (Schmidt, 1981). The final report used three assumptions to extrapolate data to full scale:

- scaling factors from the number of pins in the experiment to number of fuel pins on the HEDD's path in a full assembly,
- aerosol release fractions from the scale cask being the same as in a full scale cask, and
- a scaling factor of 5.2 to account for geometrical differences between how the test HEDD and the reference basis threat HEDD would interact to produce aerosol from affected pins

From this the BCL investigators predicted that for a full-scale test of an optimally placed HEDD charge on a truck cask carrying one 15 x 15 spent fuel pin assembly, a respirable release fraction (RRFc) of approximately $2x10^{-5}$.

Included in the BCL test results were measures of disproportionate behavior of volatile nuclides in the aerosols as a result of exposure to the high energy environment produced by an HEDD. Analysis of various aerosol size fractions demonstrated that radio-cesium was present in disproportionate amounts in the smaller size fractions examined. The degree of disproportionation in Test 3 (where enough material was sampled) was a factor of 10 in the size fraction smaller than 0.5 µm AD (aerodynamic diameter) compared to particles larger than 4 µm AD.

SNL Experimental Program: - Sponsored by the USDOE, a series of sub-scale and full-scale tests were carried out at SNL between 1980 and 1982 (Sandoval, 1983). The main thrust of the experiments was to determine what fraction of a spent fuel cask's contents could be turned into an aerosol, released, and dispersed away from the immediate vicinity of a sabotage attack using a HEDD. One full scale and two ¼ scaled tests thus were concerned with both the production of surrogate aerosols by an HEDD and also the processes that affected the fraction of aerosols that might escape to the environment in an attack.

Researchers from the Idaho National Engineering Laboratory (INEL)[2] were brought into the program to perform small scale HEDD experiments with depleted uranium pellets and actual spent fuel pellets in order to investigate differences in aerosols characteristics that might be linked to the effects of irradiation on the properties of the material. The goal of the INEL work was to measure the ratio of spent fuel aerosol mass to that of the depleted uranium surrogate aerosol mass (now referred to as the SFR, Spent Fuel Ratio). The experiments were performed and yielded information on disproportionation as well as SFR for HEDD environments. The INEL tests confirmed the BCL findings that disproportionation occurs for cesium, antimony and ruthenium, but seemed to be confined chiefly to particles 1 µm AD and smaller. No attempt to estimate the integrated effect of disproportionation over the entire respirable aerosol was attempted. Because a key set of samples were lost as a result of an accident, a reliable estimate of SFR was not produced.

In the course of the study a close working relationship developed with the program at BCL that lead to inclusion of some BCL results in the conclusions in the SNL final report (Sandoval, 1982). That report indicated that the likely release fraction (RRFc) for an HEDD attack estimated based on the available information was between $2.4x10^{-5}$ and $3.4x10^{-5}$ for 3 and 1 assembly casks, respectively. The results were based on a worst case SFR of 5.6 derived from the INEL experiments, but it was noted that other estimates (0.42, 0.53, 0.71, and 3.0) were available. Using the new estimates of RRFc, radiological consequences were estimated that indicated no early fatalities and 4 to 14 LCFs, a significantly smaller radiological impact than estimated in the Urban Studies. The result of the 4 estimates is shown in Figure 2.

[2] Now called the Idaho National Laboratory (INL)

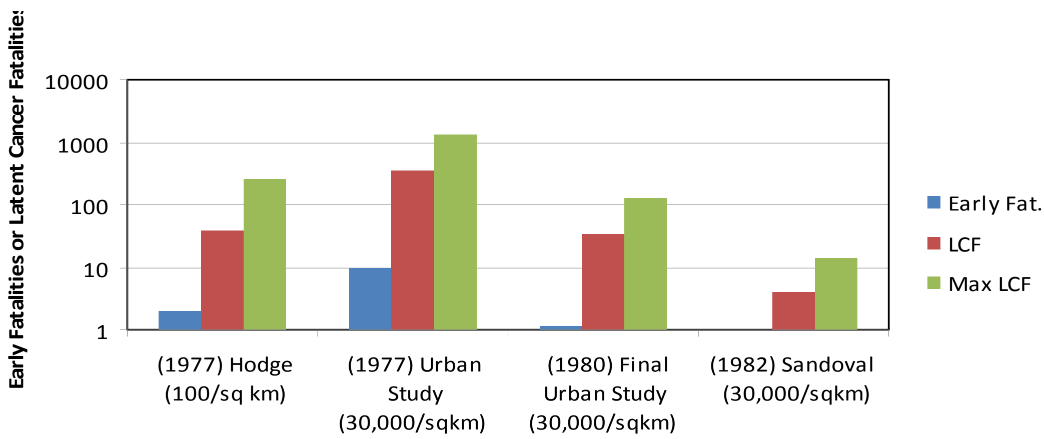


Figure 2: Estimates of Consequence of Sabotage on a 3 Assembly SF Cask

Yucca Mountain Project: **-** In 1999, The DOE commissioned SNL to make an estimate of the release fraction that might result from an HEDD attack on a typical truck or rail cask that might be used to transport spent fuel from reactors to the repository. The estimate was based on three principal parameters:

- an SRF of 3 rather than the value 5.6 used by Sandoval. The basis for using an SRF of 3 was a reexamination of data for relative post-shot concentration of aerosols from the BCL DUO_2 and spent fuel experiments that is described in the report (Luna, 1999).
- a respirable aerosol production fraction (RF) of 5% of the affected volume of spent fuel pellets that was derived from a large population of hammer tests of brittle solids.
- aerosol mass release data generated by the earlier SNL project (Sandoval, 1982).

The results of the release fraction calculation were used in the Draft YMP EIS to derive an estimate of 2 and 15 LCFs for rail and truck transport respectively. In 1994 however, data from an experimental program by GRS (Lange, 2001) was brought to light that prompted a reexamination of the YMP release estimates.

GRS Experiments: **-** The GRS projects were granted by the Federal Ministry for the Environment, Nature Conservation and Nuclear Safety (BMU) and the Federal Office for Radiation Protection (BfS). The GRS experiments were conducted at a French test site in which an essentially full scale nine assembly cask (i.e., shortened length) with pressurized DUO_2 surrogate fuel elements was subjected to 3 simulated HEDD attacks. It was intended that two of the three tests would be identical with the cask at atmospheric pressure. The third test was with the cask held at 0.8 bar, but all three used nominally the same sized HEDD. The first and second test had different damage geometries, but ended up releasing about the same amount of respirable aerosol while the third released less aerosol with the cask partially evacuated. The release amounts were lower than might have been expected applying the YMP model and the fact they were the same for different penetration geometries suggested that aerosol particles produced by the HEDD interaction behind the first surrogate fuel assembly were essentially trapped in the interior basket volumes.

Based on the GRS experiments, a revised YMP model was formulated using the earlier SNL ¼ and full scale data and the GRS results. The "GRS Model" used the GRS data from all three experiments to derive a single cask deposition factor that allowed duplication of the released mass of the three GRS experiments. Subsequently, the "GRS Model" was used to re-estimate the potential sabotage releases for the YMP final EIS (Luna, 2006) that resulted in downward revision of the release fraction estimates used in the final EIS of the Yucca Mountain Project.

Other Prior Work: **-** Early in the new millennium a paper (Ruhmann, 1985) came to light that compared the comminution behavior of DUO_2 pellets and spent fuel pellets of various low to medium burnups in severe crushing environments using a Pellini hammer apparatus. The data from the experiments yielded estimates of SFR for small particles (ca. 3-5 μm) that were in the range of 2 to 2.5 (Luna, 2004) that agree with the values produced by BCL in their experiments as analyzed for the YMP release estimate (Luna, 1999).

Building on Earlier Efforts: **-** With the large apparent variability in SFR from the SNL and BCL studies and the loss of data from the INEL experiments, an argument was made (Philbin, 2002) that the INEL experiments (or something like them) should be redone. The value for SFR varying by a factor of almost 10 from lowest to highest value meant that a consequence estimate would display the same level of uncertainty. Moreover, new experiments with surrogate materials in casks attacked with an HEDD, while valuable in the larger picture, would be devalued without persuasive confidence in a value for SFR. Although some additional information has come to light from analysis of old experiments that suggests that the SFR has a value of about 3 for spent fuel of medium burnup, a number of factors militate for additional efforts on SFR. These include:

- increase in burnup being pursued in the power industry that has significant impact on spent fuel pellet morphology (Tsai, 2003),
- the onset of long term storage in conditions not typical of earlier situations that may impact spent fuel properties, and
- decrease in radiological impact from cask accidents (USNRC, 2000) that makes the potential consequences from sabotage events the dominant radiological dose event.

The first two are related to the potential for modern practice in the fuel cycle leading to a change in the properties of the spent fuel ceramic matrix that may make it prone to different comminution mechanisms and hence different SFRs. The third issue is one that, while not technical, may be more important in justifying investment in a more reliable SFR estimate to assure the public that we understand the consequences of events that might happen that can affect them.

Overview of Recent Efforts

One constant since the late 1990's in addressing this problem has been an international working group whose primary focus has been to develop source term data from experimental simulations of sabotage-types of attacks. This working group, titled; the International Working Group for Sabotage Concerns of Transport and Storage Casks (WGSTSC) is comprised of experts mainly from the U.S., France, Germany and the UK. This group has worked to define the issues clearly and to encourage funding of research efforts in each member's country. The group has met periodically for the last decade to assess research results and point out needed efforts.

One of the projects supported strongly both directly and indirectly by the US, France and Germany has been small scale experiments at SNL that mirror the earlier works at BCL and INEL. The SNL experiments were to be conducted with greater accuracy with enough replication to yield reliable results for SFR, disproportionation and aerosol generation phenomena. In Germany, efforts were focused on aerosol production phenomenology and aerosol release from and deposition within cask-like structures. In

France emphasis has been on HEDD modeling techniques and producing appropriate analysis formalism for sabotage problems.

US Analyses and Experiments: - Starting in 2001 and continuing through 2008 an experimental program at SNL was conducted with the intent to provide a definitive measurement of the SFR. The basic concept was modeled on the 1982 INEL studies discussed above and involved the exposure of a single fuel rod segment long enough to hold 5 pellets with the high velocity jet produced by a small conical shaped charge (CSC). The entire process was to be contained in a closed volume to assure no loss of the materials generated, which was sampled to obtain information on the amounts and characteristics of the aerosols produced. Comparison of the results of identical tests with surrogate materials (DUO_2) and with actual spent fuel would provide the data from which a value for SFR can be inferred for the various types of spent fuel tested.

The test program has been extensively documented (Molecke, 2004, 2005, 2006, 2008) with special emphasis on respirable aerosol fraction for both cerium oxide and depleted uranium oxide surrogate materials. In addition, measurements of larger scale particulate and aerosol generation of other materials (Zr, Cu, Cs, Sr, Eu, Ru, and C) involved in the tests were documented. To date more than 30 tests have been completed in the process of proving containment concepts (important to authorizing experiments with spent fuel) and in gathering data on the behavior of surrogate materials in the test program. A robust test apparatus has been designed which will allow identical experiments with DUO_2 and spent fuel to be performed.

In an independent review of the results of the program (Lindgren, 2009), a re-analysis of the sampling analysis methodology and a more rigorous statistical analysis of the data from the program data resulted in changes (some significant) in the program results published earlier.
Salient results presented for respirable fractions:

- For cerium oxide surrogate pellets in the latest test series, the respirable fraction produced from the action of the CSC was stated as $6.5x10^{-3} \pm 2.3x10^{-3}$.
- For depleted uranium oxide pellets the latest test series, the respirable fraction was $1.32x10^{-2} \pm 3.2x10^{-3}$ (as modified by the re-analysis the value is $9.8x10^{-3} \pm 3.73x10^{-3}$).
- The respirable fractions are both well below the 5% value used in the YMP estimates, but the scale of these experiments is substantially smaller than those from which the 5% value was derived. The BCL program, reviewed earlier, indicated that the scaling factor between their small CSC and the reference based threat was 5.2, a value which if applied here, would bring the values obtained up to about 5%. The applicability and magnitude of the BCL scaling factor from scale lab results to real life threats needs to be resolved.
- Results for disproportionation or “enrichment” measurements based on cerium oxide pellets uniformly doped with cesium and other nuclides resulted in a value for Cs of 15±4. Measurement uncertainties for other nuclides made the results unreliable.

Extensive cooperation with French and German agencies and support in kind to the experiments has been a hallmark of this program. In addition, copious institutional problems related to safety approvals, data sharing, and disposing of the waste generated were addressed and resolved. However, as a result of program delays and funding issues, the final experimental phase with spent fuel has not been accomplished. As a result, this program has not provided the direct measurement of SFR for which it was designed.

French Analyses and Experiments: - From the years 1990, IRSN has been pursuing a multiyear program where the resistance of various casks to various threats is studied. Out of this program, based on both

experiments and numerical analysis, notably those performed in the framework of the WGSTSC, the question of estimating the release after perforation has lead to a specific approach (Loiseau, 2009). The approach developed and used at IRSN is introduced by the statement of a generic problem. Influent parameters which need to be addressed are identified. The most seducing aspect of the approach is the fact that it relies on only six parameters: the five parameters relate to (i) the energy sources capable of moving the material from the inside to the outside, (ii) the cask resistance and (iii) the release mechanisms and physics.
As an extension, most recent experiments (Autrusson, 2009) were dedicated to the study of resuspension factors of powder material in case of high energy impact. The intent was to reduce the level of uncertainty in source term derivation obtained when using resuspension factors usually considered in low energy drop or impact tests. The project has led to an improvement and a better understanding of the relevant processes and of the material properties affecting the resuspension of powder material in case of a high energy impact. These experiments have been feasible thanks to the skills and means developed by GRS and the Fraunhofer Institute which were active contributors to the latter experiments.

***German Analyses and Experiments:* -** Apart from the cooperation within the SNL and IRSN experimental programs mentioned above, GRS and the two Fraunhofer Institutes for Toxicology and Experimental Medicine (ITEM) and for High-Speed Dynamics (EMI) have conducted two experimental research projects. One of these projects concentrated on the analysis of source term generation from shaped charge impact on vitrified high level waste (VHLW). These tests were performed with non radioactive but chemically equivalent specimens of different sizes. Some of the results documented in (Brücher, 2007) are:

- Disproportionation or "enrichment" depends on the available volume for brittle fracture compared to the volume of direct CSC jet interaction with the material and therefore decreases with increasing target size.
- An upper bound for disproportionation of Cs compounds in VHLW is 10.
- The observed aerosol source term supports estimates from former Battelle experiments with inactive glass specimen.

In 2008, a new test program was started with the aim of analyzing particular phenomena and parameters affecting the primary (internal) or the secondary (outside) source term of a CSC attack on a transport and storage cask for spent fuel or other radioactive material. Parameters of interest are, inter alia, the source term fraction from different inventory locations, the primary source term distribution inside the cask and the influence of initial pressure on the release from the cask. The test program includes laboratory scale experiments as well as full scale experiments to verify the transferability of results between laboratory scale and real scale.

The final objective of the project is to develop and validate a numerical model chain which simulates the relevant processes of penetration, primary source term generation and release to the environment for arbitrary configurations of packaging and radioactive inventory. Still ongoing, the test program has already led to consistent and repeatable results in the laboratory scale; e.g. a rather small but still detectable outside source term even if the cask atmosphere pressure remains below ambient conditions. These results will allow to further refine and to improve the model system and to prepare the following full scale test phase.

Conclusion

The potential consequence of a sabotage attack on a spent fuel shipment has been a concern to governments and the public since the mid-1970s. The level of concern has translated into a number of experimental, analytical, and institutional actions over the ensuing years which have been reviewed above. Based on these programs and actions, a level of understanding has been achieved as outlined below:

- Because of their robust construction and performance based design, spent fuel casks are unlikely to release a significant fraction of their content in most realistic sabotage scenarios.
- One full scale test using realistic HEDDs on casks initially at sub-atmospheric pressure and containing surrogate spent fuel elements has produced releases about 60% lower than unpressurized casks. This test, as well as recent laboratory scale results suggest that initial over/under pressurization is an important amplifying/ameliorating factor in releasing respirable material.
- A number of lab scale experiments with spent fuel surrogate and actual medium burnup spent fuel subjected to high energy environments have indicated that the mass of respirable spent fuel aerosol to surrogate material aerosol (the SFR) for the same experimental conditions has a value that is about 3.
- A relatively higher concentration of volatile nuclides (relative to their bulk concentration) in aerosols from lab scale experiments with real spent fuel and doped surrogates has been observed in the respirable fraction of aerosols. The extra amount, termed disproportionation or enrichment is a factor of 10 to 20 above what would be expected based on gross specie concentration.
- A heuristic/empirical model for releases from spent fuel casks subjected to HEDD attack has been developed based on the two existing fuel scale experiments. More full scale experiments are needed to understand aerosol affecting processes occurring within the cask in order to develop a more omnibus model.
- Respirable surrogate aerosols from small scale experiments indicate a respirable fraction from the action of a CSC is approximately 1%. Scaled to a reference based threat using a factor developed by BCL would indicate a full scale respirable fraction of about 5%. The applicability and magnitude of the BCL geometric factor in these cases needs to be resolved.

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