Proceedings of INMM 57th Annual Meeting
Atlanta Marriott Marquis, Atlanta, GA
July 24-28, 2016

SAND2016-5419C

Reexamination of Respirable Release Fraction Data for Spent Nuclear Fuel

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ABSTRACT

Sabotage of spent nuclear fuel casks remains a concern nearly forty years after attacks against shipment casks were first analyzed. A limited number of full-scale tests and supporting efforts using surrogate materials, typically depleted uranium dioxide (DUO_2), have been conducted in the interim to more definitively determine the source term from these postulated events. Additional small scale studies have been conducted to characterize the aerosols generated from the high-energy disruption of fresh, irradiated, and surrogate fuel samples. These efforts presented respirable release fraction (RRF) data as a function of energy density. The respirable release fraction is one of the principal quantities that defines the source term and directly relates to the consequence of an event. A thorough understanding of the relevant RRF data is therefore required for best-estimate consequence analyses and statistical interpretations. Previous examinations of available fragmentation data involving spent nuclear fuel (SNF) and surrogates have been presented in two extensive handbooks, one published by the Nuclear Regulatory Commission (NRC) and the other by the Department of Energy (DOE). These handbooks present RRF data with little discussion of the underlying assumptions in their interpretations. The current analysis will reconstruct the existing handbook results from the source data and present alternative assumptions and interpretations, which are statistically defensible. The RRF presented in the NRC handbook gives values that are approximately 3 to 10 times higher than the best-estimate (least-squares) values derived directly from the original data over the energy densities of interest.

INTRODUCTION

Previous examinations of available fragmentation data involving spent nuclear fuel (SNF) and surrogates have been presented in two reference documents [NUREG/CR-6410 and DOE HDBK-3010-94]. These references present this data with little discussion of the underlying assumptions in their interpretations. The current analysis will reconstruct the existing reference results and present alternative assumptions/interpretations, which are more defensible in the opinion of the authors.

HIGH-ENERGY DENSITY DATA ANALYSIS

The data analyzed for the high-energy density range includes aerosol samples from spent fuel collected at the Kraftwerk Union AG (KWU) [Ruhmann, et al. 1985]. Only the 33 GWd/MTHM samples from Ruhmann were included in NUREG/CR-6410. However, the 0 and 22 GWd/MTHM samples are also presented for the current analysis.

Figure 1, Figure 2, and Figure 3 show the cumulative mass distributions for 33, 22, and 0 GWd/MTHM samples [Ruhmann, et al. 1985]. The original graphs have been inserted as a background image for the reader's convenience. The corresponding log-normal distributions are presented for each data set. All curve fits presented in this paper are best estimate based on least-squares (LSQ) regressions. Each data set was collected for a different energy density input (e.g. E_d = 1000 J/g). Some of the data points in the original graphs were shifted to provide clarity. However, the digitized points were placed at the reported mesh sizes for which the data was collected. The mass median diameter and geometric standard deviation are presented in the legends.

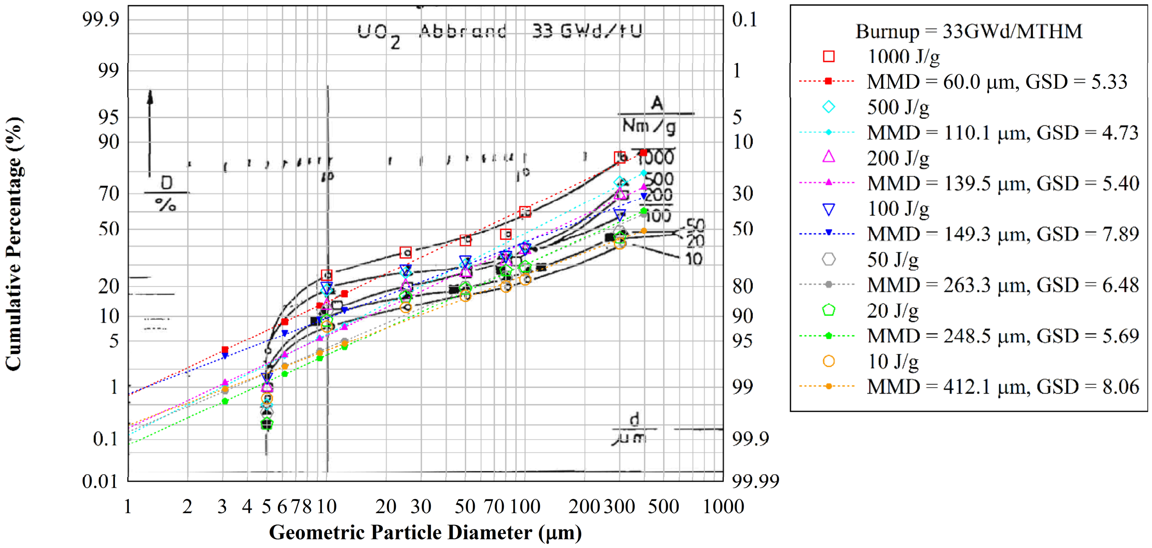

Figure 1 **Cumulative mass distribution curves for fragments generated from SNF with a burnup of 33 GWd/MTHM [Ruhmann, et al. 1985].**

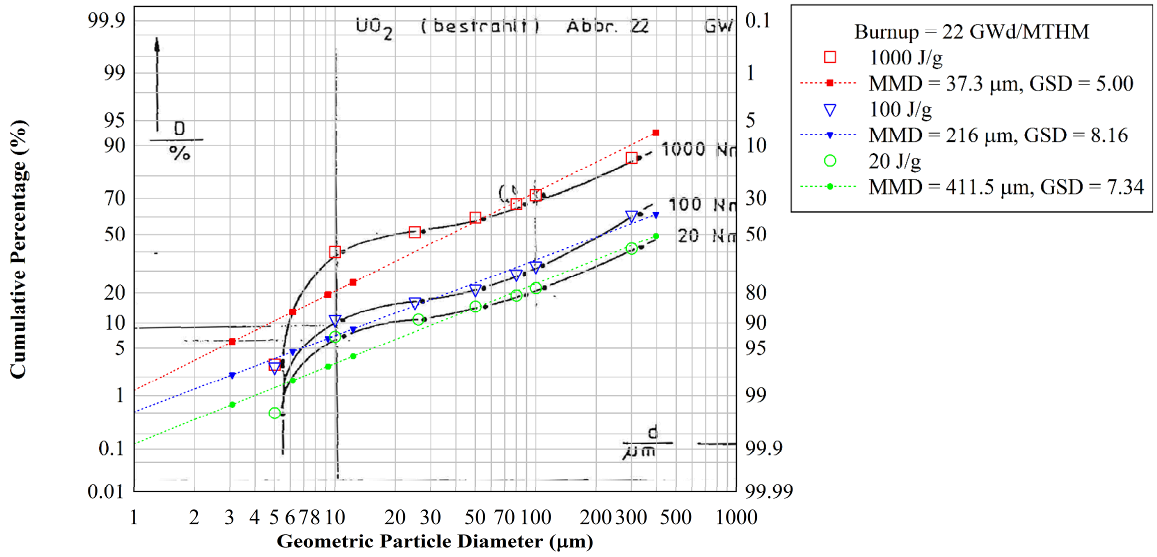

Figure 2 Cumulative mass distribution curves for fragments generated from SNF with a burnup of 22 GWd/MTHM [Ruhmann, et al. 1985].

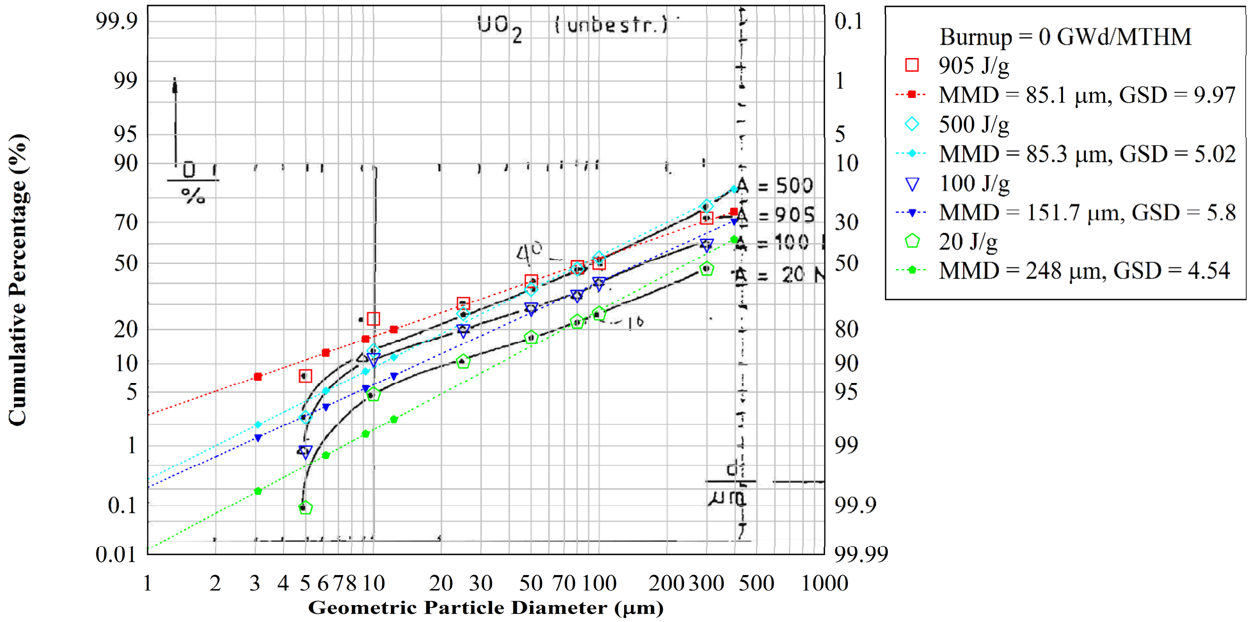

Figure 3 Cumulative mass distribution curves for fragments generated from unirradiated fuel [Ruhmann, et al. 1985].

NUREG/CR-6410 presents the following relationship for the aerodynamic mass median diameters (AMMD) in µm for the 33 GWd/MTHM samples as a function of energy density in J/m^3.

$$\text{AMMD} = 5842 - 557.8 \log(E_d) \qquad (1)$$

The given equation for AMMD is plotted against available data in Figure 4. The curve fit given in NUREG/CR-6410 yields values that are on average 42% higher than the currently derived curve fit to the data. Ultimately, this would result in respirable release fractions lower than calculated in the current analysis. However, the next assumption in NUREG/CR-6410 states, “the data have an average geometric standard deviation, σ_g (GSD in this report), of 19.” It is unclear how this assumption was derived or the justification for it. Assuming a value of GSD = 19 for distributions that have an arithmetic average of GSD = 6.4 appears to be indefensibly conservative.

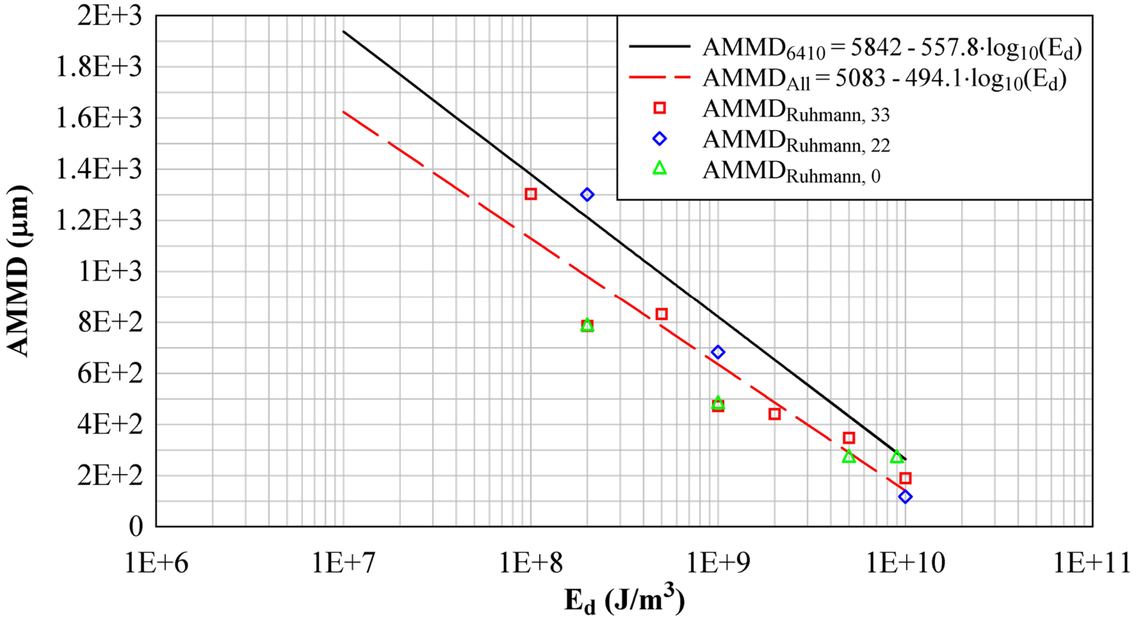

Figure 4 **Aerodynamic mass median diameter as a function of energy density for all data.**

Figure 5 shows the respirable release fraction (RRF), i.e. the percent less than 10 μm AED, as a function of energy density. The values in NUREG/CR-6410 are reproduced along with the available Ruhmann data. The Ruhmann data points were derived by using the AMMD and GSD for each sample set. As can be seen in the plot, the available data points give values much smaller than those indicated in NUREG/CR-6410.

$$\mathrm{RRF} = 0.5\left\{1+\mathrm{erf}\left[\ln(10\ \mu\mathrm{m}/\mathrm{AMMD})/\left(2^{0.5}\ln(\mathrm{GSD})\right)\right]\right\} \qquad (2)$$

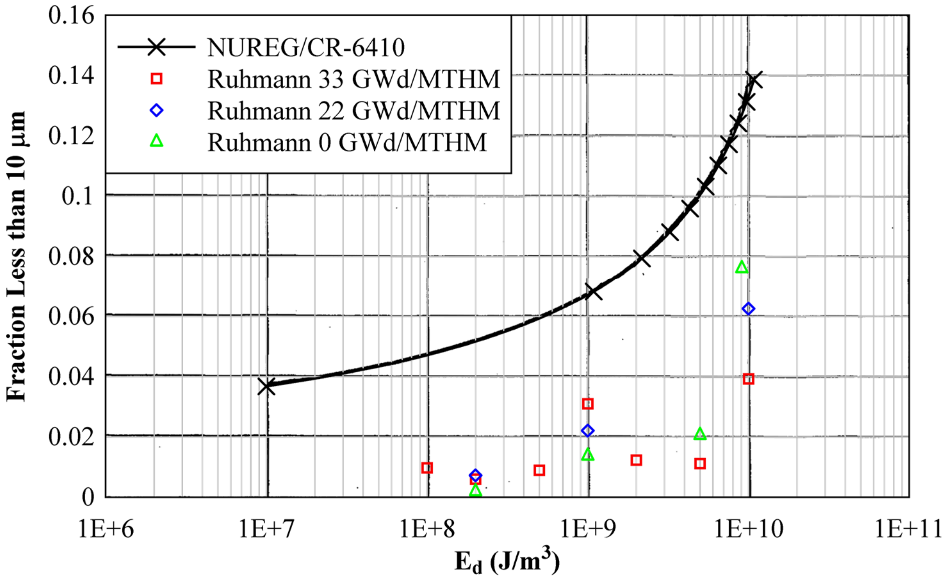

Figure 5 **Respirable release fraction as a function of energy density plotted on a log-linear scale [NUREG/CR-6410].**

DOE-HDBK-3010-94 presents the following curve fit for the respirable release fraction as of energy density in J/m^3. It is assumed that the RRF plateaus after reaching a value of 1.

$$RRF_{DOE} = 2E\text{-}10\ (E_d) \tag{3}$$

Figure 6 shows the same data as in Figure 5 but now in log-log space. A curve fit to all the Ruhmann data is presented. The prediction bands for the complete Ruhmann data set are shown as shaded regions about the RRF_{All} curve fit. These prediction bands give the range over which 95% of possible data could exist based on the current data set. The NUREG/CR-6410 data fit most closely corresponds to the upper prediction bands of the two curve fits. However, no description was found in NUREG/CR-6410 that indicates that the RRF was taken to be at the upper statistical bounds of the data. The DOE correlation is also shown for reference.

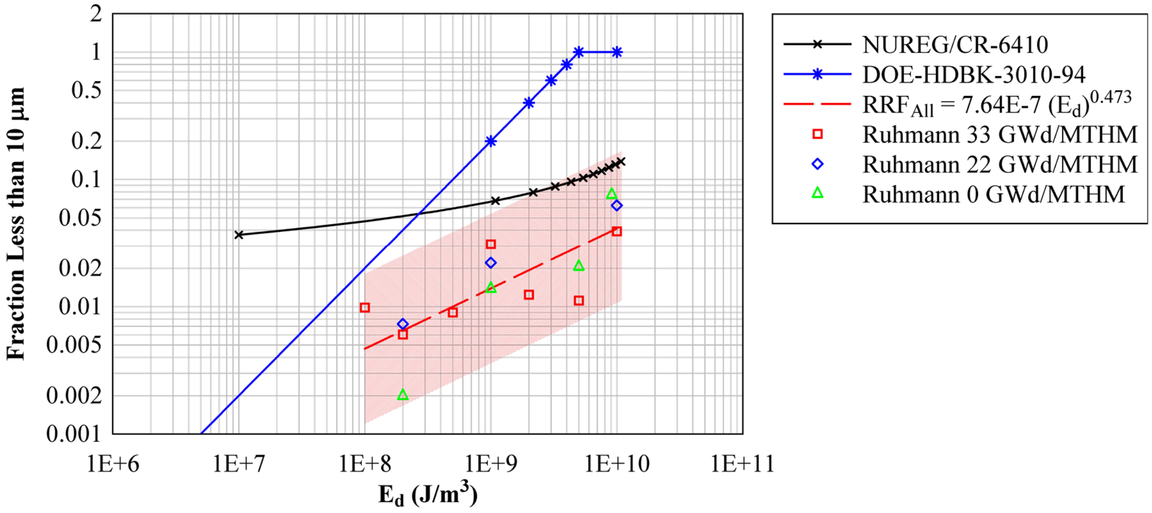

Figure 6 Respirable release fraction as a function of energy density plotted on a log-log scale.

Figure 7 shows the ratio of the RRF found in NUREG/CR-6410 to the RRF calculated from all of the Ruhmann data for a least-squares fit and the 95% prediction bands. The RRF in NUREG/CR-6410 gives values that are approximately 10 to 3 times higher than the best estimate (least-squares) values derived directly from the Ruhmann data over the energy densities of interest. Assuming that the least-squares regression of the Ruhmann data offers the best estimate of RRF, the curve fit for RRF offered in NUREG/CR-6410 appears to be overly conservative.

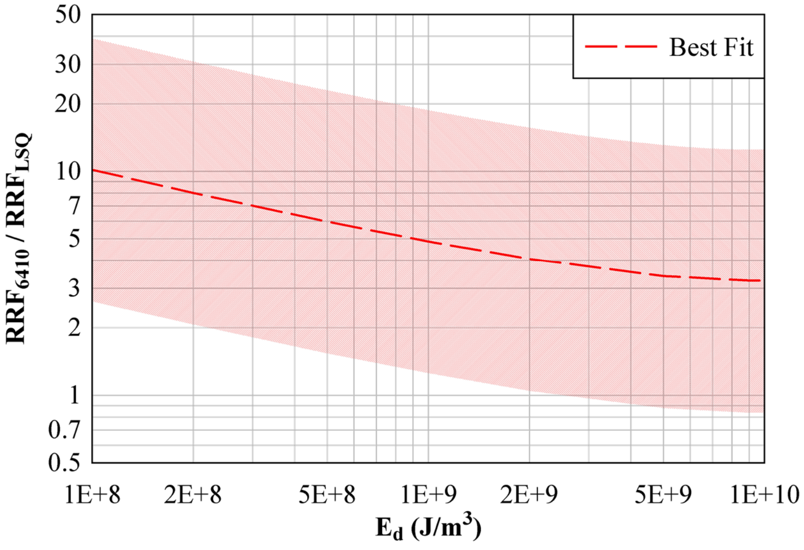

Figure 7 Ratio of respirable release fractions from NUREG/CR-6410 to the least-squares (dashed red line) and prediction bands (red shading) for the Ruhmann data.

LOW-ENERGY DENSITY DATA ANALYSIS

The data analyzed for the low-energy density range includes aerosol samples from several different brittle glasses and ceramics under consideration for waste forms at Argonne National Laboratory (ANL) [Jardine, et al. 1982]. The original data and the correctly calculated respirable release fractions are presented in Table 1. The average data for a mass fraction less than 10 µm geometric is approximately 2.6 times higher than the corresponding mass fraction for particles less than 10 µm AED. Note that Jardine reports respirable fractions at 10 µm geometric diameter regardless of sample density. The current analysis correctly accounts for the density in the calculation of the respirable fraction.

Table 1 Original data from Jardine, et al. 1982 and calculated respirable release percentages.

Sample Type	Table Origin	Mass Median Diam. (mm)	GSD	Reported <10 µm Geo. (%)	Calulated <10 µm Geo. (%)	Calulated <10 µm AED (%)	Geo./AED
SRL 131 (1)	6	2.6	6.4	0.14	0.14	0.05	2.6
SRL 131 (2)	6	2.6	6.6	0.18	0.16	0.07	2.5
High Silica	6	3.7	8.5	0.29	0.29	0.14	2.0
Alkoxide	6	2.2	7	0.27	0.28	0.13	2.1
PNL 76-68	6	2.3	6.5	0.17	0.18	0.07	2.6
Pyrex	6	1.4	6.0	0.27	0.29	0.14	2.0
Pyrex	9	0.18	4.7	3.2	3.09	1.68	1.8
SRL 131	9	0.32	5.2	1.7	1.78	0.80	2.2
SRL 131	7	2.7	6.8	0.16	0.17	0.07	2.4
SRL 131	7	5.4	6.1	0.087	0.03	0.01	2.9
SRL 131	7	5.0	7.5	0.031	0.10	0.04	2.4
SRL 131	7	9.5	6.7	0.016	0.02	0.01	2.9
Pyrex	7	1.7	6.3	0.27	0.26	0.13	2.0
Pyrex	7	3.4	6.7	0.11	0.11	0.05	2.1
Pyrex	7	6.9	7.3	0.052	0.05	0.02	2.1
Pyrex	7	11	8.7	0.067	0.06	0.03	1.9
SYNROC B	6	4.2	7.6	0.15	0.14	0.04	3.3
SYNROC D	6	4.7	8.1	0.16	0.16	0.05	3.0
SYNROC C ANL (1)	6	6.4	8.2	0.15	0.11	0.03	3.4
SYNROC C ANL (2)	6	10	9.6	0.13	0.11	0.04	3.0
Tailored	6	13.7	9.3	0.06	0.06	0.02	3.1
SYNROC B	9	0.59	5.4	0.76	0.78	0.23	3.5
SYNROC D	9	0.52	5.9	1.2	1.30	0.45	2.9
SYNROC (RAMM)	10	0.41	4.8	0.9	0.90	0.24	3.7
SYNROC (RAMM)	10	0.65	7.3	1.8	1.79	0.71	2.5
SYNROC (RAMM)	10	0.70	7.4	1.8	1.69	0.67	2.5
FUETAP	6	2.3	7.9	0.43	0.43	0.28	1.5

NUREG/CR 6410 presents the following curve fit for the respirable Jardine data as a function of energy density in J/m^3. This curve fit is recommended for energy densities less than 1E+07 J/m^3.

$$RRF_{6410} = 3.27E\text{-}11\ (E_d)^{1.131} \tag{4}$$

Figure 8 shows the low-energy density data for the as-reported respirable and as-calculated respirable fractions from Jardine. The recommended curve fits for respirable release fractions (RRF's) from NUREG/CR-6410 and DOE-HDBK-3010-94 are also shown in the figure. It is unclear from the NUREG/CR which exact data from Jardine was used to construct the curve fit. The curve fit may have been constructed using a subset of available data. New power law fits through the geometric and AED data are shown in the plot. The suggested exponent of 0.784 is lower than purported in NUREG/CR-6410. The prediction band about the AED curve fit is represented as a shaded band.

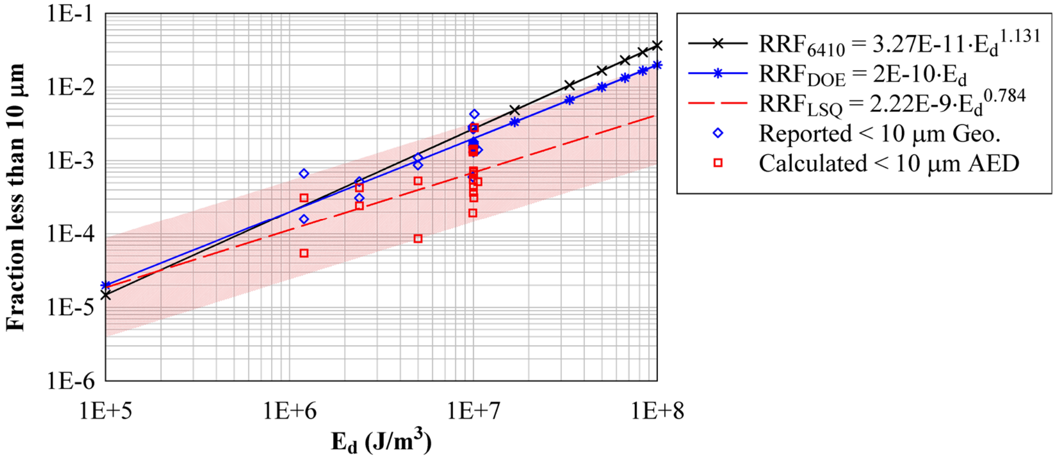

Figure 8 **Low-energy density respirable data.**

CONCLUSIONS

The underlying data used to derive the curve fits for respirable release fractions from NUREG/CR-6410 and DOE-HDBK-3010-94 have been reexamined to independently determine new statistically-based relationships. These investigations revealed that the original interpretations make undocumented and often highly conservative assumptions. The respirable release fraction as a function of energy density is shown for all three analyses in Figure 9. As summarized in Figure 9, the current analyses and those from NUREG/CR-6410 use the data from Jardine, et al. and Ruhmann, et al. to determine the RRF in the low-energy and high-energy ranges, respectively. The current least-squares (LSQ) analyses show that the RRF is overestimated in the previous treatments at all but the lowest end of the energy density range as shown in Figure 10. For the range of the middle to the upper energy densities, the integrated average RRF is less than a factor of approximately four compared to the correlations given in NUREG/CR-6410. For the same range, the DOE correlation gives an integrated average of RRF 30 times higher than the current analyses.

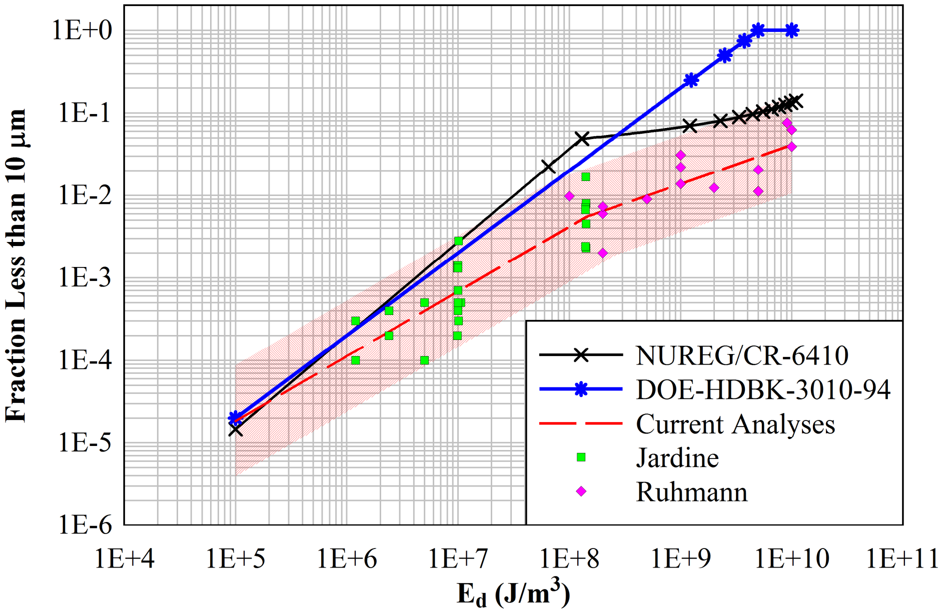

Figure 9 Respirable releases calculated with different methods for low and high-energy density ranges.

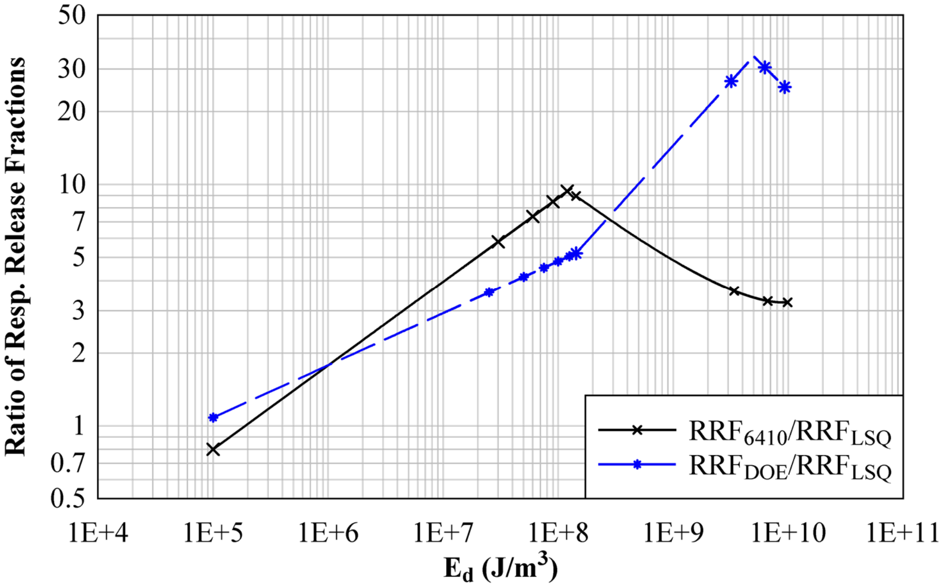

Figure 10 Respirable release fractions from NUREG/CR-6410 and DOE-HDBK-3010-94 normalized to the currently derived values as a function of energy density.

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