

## **AEROSOL GENERATION FROM SPENT FUEL IN HIGH ENERGY IMPACT ENVIRONMENTS: SEARCHING FOR BETTER DEFINITION OF THE ELUSIVE SFR<sup>1</sup>**

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### **ABSTRACT**

This paper reviews the work performed to date to obtain a better definition of the spent fuel to surrogate aerosol ratio, which is of prime interest in evaluating the effects of high energy environment on the behavior of spent nuclear fuel in accidents and potential terrorists' attacks. It is necessary to have information on the particulate produced within the cask as a result of high-energy impact environments. In the event of a leak path to the environment, the particulate released to the cask cavity is required in order to estimate the characteristics of the source term that might result in nearby persons receiving a radiation dose. This paper examines data by Ruhmann and other authors relating to the particulation of irradiated fuel and un-irradiated UO<sub>2</sub> for identical experimental conditions and its application to estimating a value for SFR.

### **INTRODUCTION**

Evaluating the effects of high energy environments on the behavior of spent nuclear fuel is of prime interest in evaluating the effects of severe accidents involving spent fuel casks. In addition, similar impact environments can result from effects of potential terrorist acts involving spent fuel casks. In both cases it is necessary to have information on the particulate produced within the cask as a result of high-energy objects impacting spent fuel rods. In the event the impact also produces a leak path to the environment, the quantity of particulate released to the cask cavity is required in order to estimate the characteristics of the source term for producing radiological dose in nearby persons. Of particular interest are particles that are respirable (those with diameter < 10 micrometers aerodynamic diameter<sup>2</sup> AD). A review of earlier work in this area (Luna, 1999) suggested that the ratio of the respirable mass of spent fuel particles to those produced from a similar experiment on unirradiated UO<sub>2</sub> fuel pellets, termed the SFR, was approximately 3. However, it was noted in that analysis that the established values ranged from less than 1 to about 10. In such a situation, additional data that might yield a narrowing of the range would be of value in making better estimates of the potential radiological consequences from an attack. As a result, efforts have been underway since 2000 to develop a new set of experiments to measure the SFR in hopes of narrowing the uncertainty band on this parameter particularly for spent fuel with burnups greater than 33 Gwd/tU expected to be the predominate spent fuel discharged from reactors in the future. Progress reports relating to development of these experiments have been presented at previous Waste Management Symposia, PATRAM,

HLRWM and INMM conferences (Lake, 2001; Lake, 2002; Luna, 2001, Molecke 2003a, Molecke 2003b). These papers describe a multinational test program that will quantify the aerosol particulates produced when a high energy density device (HEDD) impacts surrogate material test rods and actual spent fuel test rods. This spent fuel sabotage - aerosol test program is coordinated with the international Working Group for Sabotage Concerns of Transport and Storage Casks (WGSTSC) and supported by both the U.S. Department of Energy and Nuclear Regulatory Commission. The WGSTSC has international organization participants from the U.S., Germany, France, and Great Britain.

## **ANALYSIS OF RUHMANN EXPERIMENTS AND DATA**

Not reviewed in the above report (Luna, 1999) was a report by Ruhmann and other authors (Ruhmann, 1985) containing data relating to the particulation of irradiated fuel and un-irradiated  $\text{UO}_2$  in identical experimental conditions. The report described experiments conducted with un-irradiated  $\text{UO}_2$  and irradiated spent fuel pellets with burn-ups of 33 Gwd/tU and 22 Gwd/tU. The experiments were done in an apparatus usually referred to as a Pellini hammer. Basically a heavy weight is dropped on a pellet or pellet fragments in a contained space from a range of heights to give various energy inputs per mass or volume of impacted material. The particulate material so produced was collected and sieved to obtain data relating particle geometric diameter and mass in a diameter range for the test.

A similar apparatus was used by Jardine in an earlier study (Jardine, 1982) to obtain estimates of the particulate produced when various glasses, proposed high level waste disposal forms, cements and  $\text{UO}_2$  pellets were subjected to impact environments of varying severity (as measured by the energy imparted per unit volume of material). Jardine fit lognormal distributions to the sieve results and then plotted the particle mass fraction smaller than 10 micrometers geometric diameter<sup>3</sup> versus energy per unit volume. The relationship obtained indicated that the less than 10-micrometer fraction was related to energy density by a power-law relationship (with  $n \approx 1$ ). In addition, the results indicated that the mass median diameter of the lognormal fit decreased almost linearly with energy density and the geometric standard deviation was almost constant with energy density (decreased slowly with energy density).

Although Ruhmann used sieves yielding mass versus diameter data down to a 5 micrometer size, the Ruhmann data is probably only reliable down to about 10 micrometers geometric diameter. The 5-micrometer data is probably unreliable because, as pointed out by Ruhmann in his paper, the initial grain size of the spent fuel and  $\text{UO}_2$  was in the 5 to 10 micron range and a different fracture mechanism is likely to govern breakage of the feed material grains compared with breakup along grain boundaries. Since the respirable size threshold at 10 micrometers AD relates to a geometric diameter of about 3 micrometers, the SFR estimates derived from Ruhmann's data are not quite what is needed, but the data indicate the general trends for SFR as particle size approaches the respirable limit.

## **SPENT FUEL TO SURROGATE AEROSOL RATIO**

The process used in this paper for obtaining SFR estimates from the Ruhmann data was as follows:

- Data points on the mass less than diameter,  $d$ , versus  $d$  curves for each energy loading (J/g) in the Ruhmann report were read off as accurately as possible for all the experiments (33 Gwd/tU spent fuel, 22 Gwd/tU spent fuel, and unirradiated  $UO_2$ ). The data are provided in Figures A1, A2, and A3.

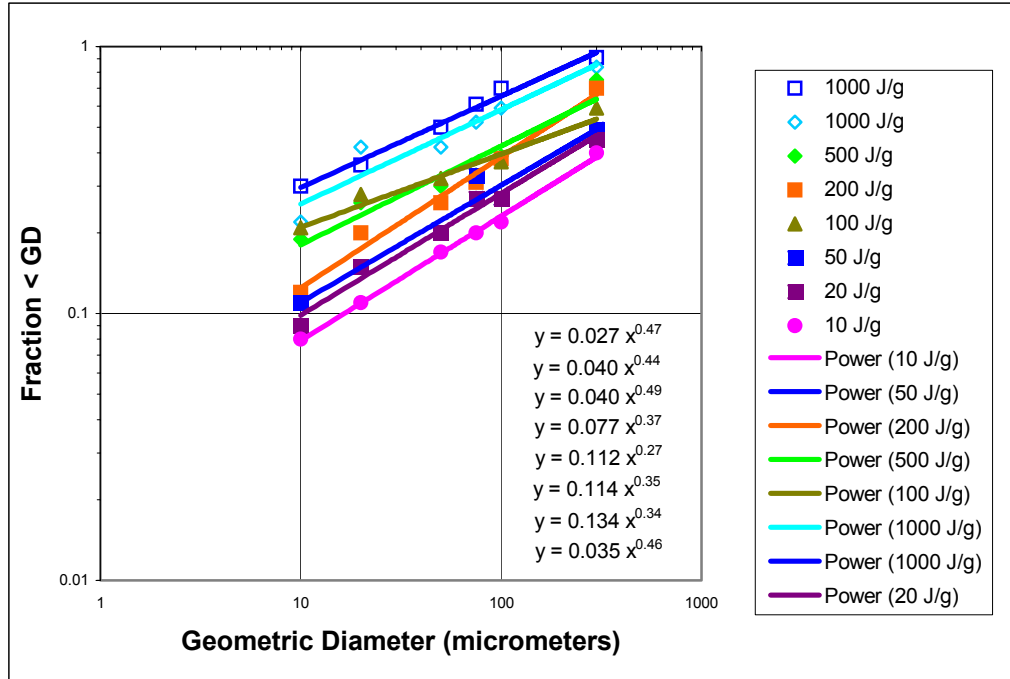


Fig. A1 Ruhmann particle size data for 33 Gwd/tU irradiation

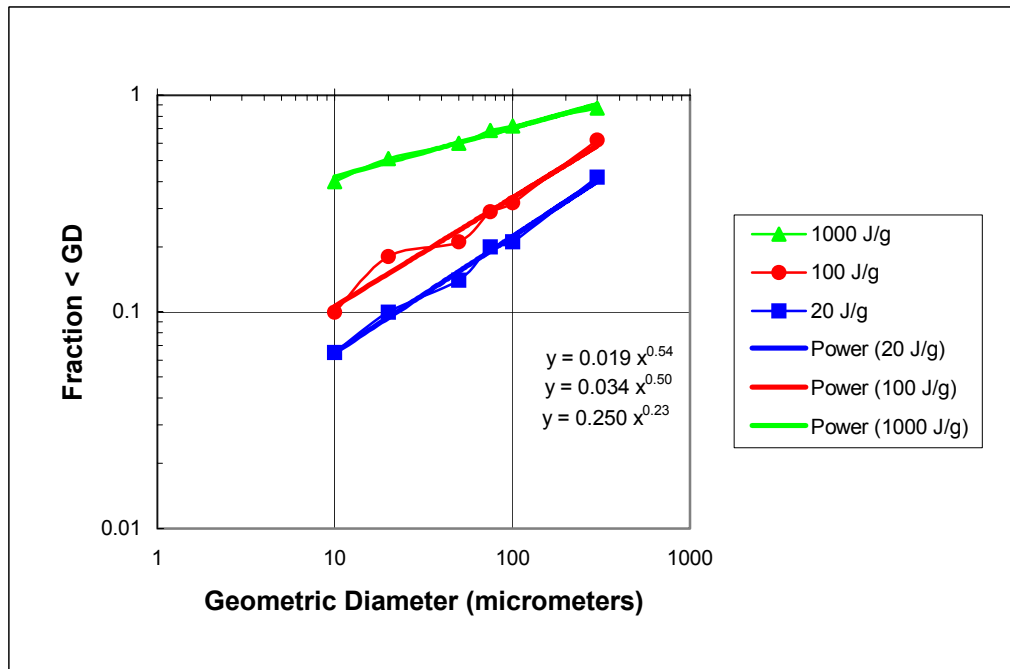


Fig. A2 Ruhmann particle size data for 22 Gwd/tU irradiation

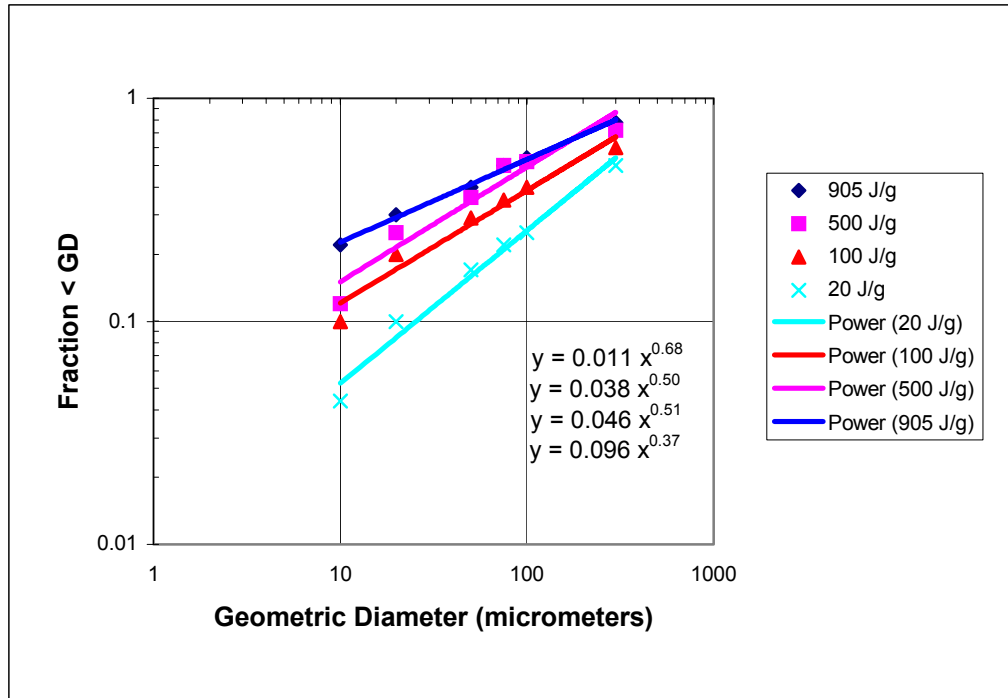


Fig. A3 Ruhmann particle size data for unirradiated UO<sub>2</sub>

- The data were transferred to an EXCEL spreadsheet for analysis.
- For each loading and burnup combination, the ratio of spent fuel mass smaller than d to unirradiated UO<sub>2</sub> mass smaller than d was calculated. These data are shown in Table I.

Table I SFR Estimates Derived from Ruhmann, et al, 1985

geometric diameter in (micrometers)	(SFRs) Ratio of Irradiated to Unirradiated Mass Fraction smaller than d [for indicated Loading (J/g) / Burnup (Gwd/tU)]								Averaged Values for Given Burnup (Gwd/t)		
	1000/22	100/22	20/22	1000/33	1000/33	500/33	100/33	20/33	Avg. 22	Avg. 33	Avg. All
300	1.12	1.03	0.84	1.17	1.08	1.04	0.98	0.90	0.98	1.03	1.01
100	1.33	0.80	0.84	1.30	1.09	0.75	0.93	1.08	0.94	0.99	0.97
75	1.38	0.83	0.91	1.22	1.04	0.66	0.94	1.23	0.99	0.97	0.98
50	1.50	0.72	0.82	1.25	1.05	0.83	1.10	1.18	0.92	1.06	1.00
20	1.70	0.90	1.00	1.20	1.40	1.04	1.40	1.50	1.11	1.29	1.21
<b>10</b>	<b>1.82</b>	<b>1.00</b>	<b>1.48</b>	<b>1.36</b>	<b>1.00</b>	<b>1.58</b>	<b>2.10</b>	<b>2.05</b>	<b>1.35</b>	<b>1.50</b>	<b>1.44</b>
5	0.56	3.26	1.31	1.14	1.21	0.39	1.26	1.22	1.05	0.85	0.91

As can be seen from Table I, the SFR values for relatively large diameters are approximately unity, but that for smaller diameters the SFR for most combinations of loading and burnup increases to values between 1.5 and 2.1 for the data at 10 micrometers.

If the data in this table are plotted, the curves in Figure 1 are obtained. These curves clearly show the trend to increasing SFR as diameter decreases. If the curves in Figure 1 were extrapolated to respirable sizes (3 micrometers), the indicated SFR likely would be in the range of 2 to 3. Thus the Ruhmann data tends to confirm the validity of using the SFR value of 3.

Even in light of the uncertainty in extrapolating the curves to the respirable size range, it seems clear that the value as high as 5.6 used in an earlier estimate of sabotage source term (Sandoval, 1983) was too high. This analysis also supports the use of 3 as the value for SFR that was used in assessing the likely source term for a terrorist attack on a spent fuel cask in transit to the proposed Yucca Mountain Repository (Luna, 1999; DOE 2001).

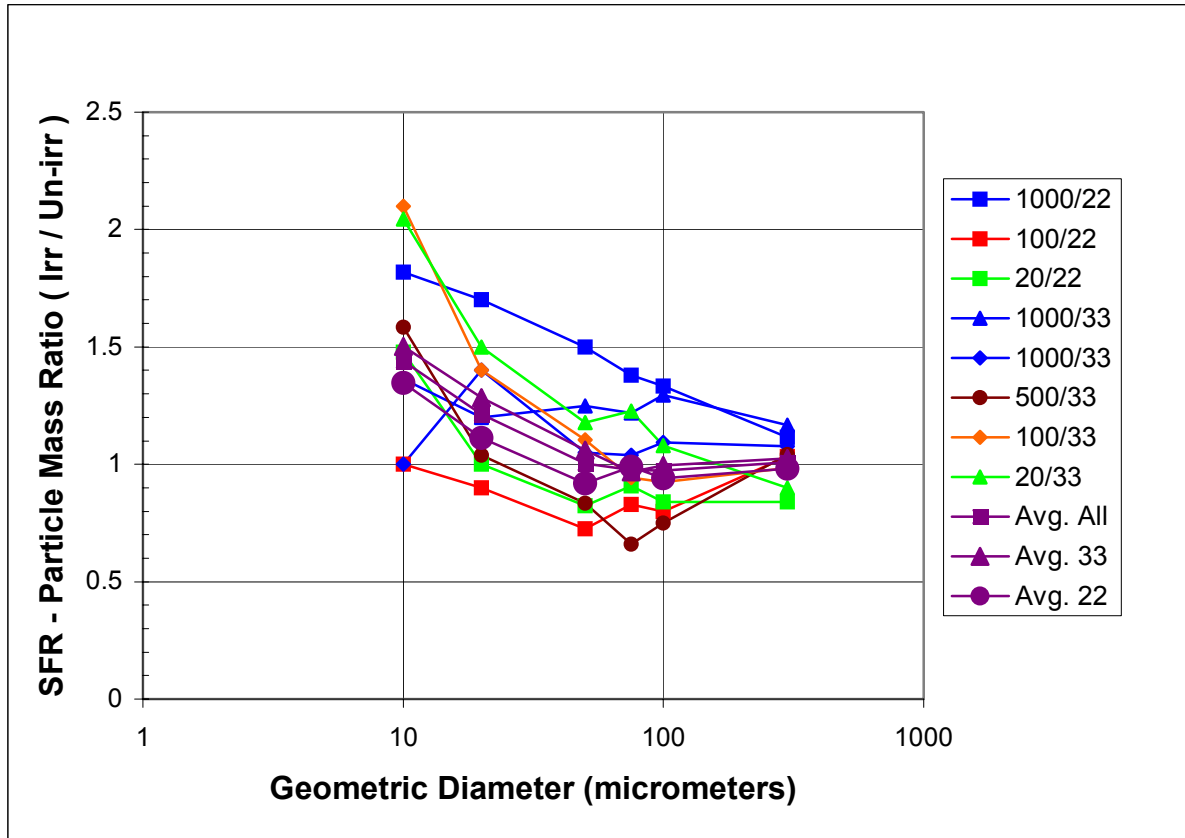


Fig. 1 SFR Values Versus Geometric Diameter, d, for Various Values of Loading / Burnup

### CONCLUSION

This analysis of the Ruhmann data together with previous data indicates that the SFR for respirable particles would be in the range of 2 to 3. These data tend to confirm the validity of using the SFR value of 3 for assessing the likely source term for a terrorist attack on a spent fuel cask being transported to the proposed Yucca Mountain Repository. While there is some uncertainty in extrapolating the curves to the respirable size range, one can conclude that a value as high as 5.6, as used in an earlier estimate of sabotage source term (Sandoval 1983), was too high. The current work being conducted by the WGSTSC will produce experimental data for validating the SFR for particulates in the respirable size range (i.e., <10 micrometers).

**REFERENCES**

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- 10 **Sandoval, 1983** – “An Assessment of the Safety of Spent Fuel Transportation in Urban Environs,” R. P. Sandoval, et al, SAND82-2365, Sandia National Laboratories, June 1983.

**FOOTNOTES**

- <sup>1</sup> SFR is the ratio of spent fuel aerosol mass to UO<sub>2</sub> aerosol mass from an impact-type event under identical (or nearly so) conditions. Usually the SFR is of primary concern for respirable particles.
- <sup>2</sup> For particles such as UO<sub>2</sub>, with a density of approximately 10 g/cc, respirable particles will have diameters smaller than 3 micrometers

- <sup>3</sup> Jardine refers to the less than 10-micron fraction as “respirable”, but with materials with densities of 2 to 3 the limiting diameter for respirability would be 6 to 7 microns.

## **ENDNOTES**

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