CLM-P816

The particle size distribution of solidified melt debris from molten fuel-coolant interaction experiments

D. F. Fletcher

UK ATOMIC ENERGY AUTHORITY

Culham Laboratory

CLM-P816

The particle size distribution of solidified melt debris from molten fuel-coolant interaction experiments

D. F. Fletcher

UKAEA, Culham Laboratory,
Abingdon, Oxon, OX14 3DB, U.K.

SUMMARY

The statistical distribution of debris from molten fuel-coolant interactions (MFCIs) is examined. It is shown that a wide range of fragmentation processes, including MFCIs, produce debris which has an upper limit lognormal (ULLN) distribution. Fitted distributions are presented for 20 experiments using 24 kg of uranium dioxide/molybdenum melt quenched in water.

(To be submitted for publication in Nuclear Engineering and Design)

Culham Laboratory
United Kingdom Atomic Energy Authority
Abingdon
Oxfordshire OX14 3DB

March 1987

1. Introduction

A knowledge of particle size distributions is of importance for many aspects of reactor severe accident analysis, including the study of steam explosions (also known as vapour explosions and MFCIs), debris bed coolability and steam spike phenomena [1]. In this paper we discuss the form of the particle size distribution appropriate to melt which has been fragmented in molten fuel-coolant interactions (MFCIs). A large amount of data is available for various 'fuel' and 'coolant' pairs, on 'fuel' mass scales ranging from a few milligrams to tens of kilogrammes. Many investigators, see for example [2], have noted that all the debris distributions take a very similar form when the distribution function is plotted on a log/probability scale. In this paper it is shown that this distribution is upper limit lognormal (ULLN).

In section 2 we describe this distribution and discuss its properties. In section 3 we describe two series of experiments carried out in the Molten Fuel Test Facility (MFTF) at AEE Winfrith. Fitted distribution parameters for approximately twenty experiments involving uranium dioxide/molybdenum melt and water are presented. Debris data from both steam explosions and slow non-explosive events has been examined.

2. The Statistical Distribution of the Debris

For many purposes it is useful to be able to describe the debris data in terms of a statistical distribution. It is well-known [3,4] that successive independent fragmentation processes lead to lognormal size distributions of the smaller fragments. This result is a consequence of the multiplicative analogue of the Central Limit Theorem of Statistics, i.e. the product of independent random variables is distributed lognormally. Of course, a lognormal distribution is not fully realisable in practice since it predicts a non-zero probability density function over the range zero to infinity. In any real situation a largest particle size is set either by the finite mass of material considered in the sample or by hydrodynamic instabilities which set a maximum stable droplet size.

A lognormal distribution gives a straight line when its cumulative distribution function is plotted on log/probability paper, whereas an upper limit lognormal distribution gives a straight line for small particle sizes, tending to an asymptote at the maximum size $x = x_m$. This distribution has been found to apply to many particle size distributions including:-

(i) Droplets produced by a spray [5].

(ii) Powders produced by crystalisation [6].

(iii) The particle size distribution for pit-sand given in [2] (fitted by the present author).

(iv) A variety of molten fuel-coolant interaction debris distributions; ranging from small drops where x_m is typically 0.2-2 mm (see figure 12 of [2]) to 4 kg quantities of UO_2 quenched in sodium (see figure 11 of [2]) where x_m is of the order of 10 mm. In the sodium case this distribution can be produced by a combination of rapid fragmentation and chemical attack. Chemical attack may occur if the fuel is oxygen rich or if methanol is used to separate UO_2 from sodium. The task of physically separating the fuel from the sodium can also cause further fragmentation.

(v) 24 kg quantities of uranium dioxide/molybdenum melt quenched in water (reported in section 3 of this paper).

In an earlier paper [7] the author carried out a detailed study of particle size distributions of debris produced from MFCI's involving UO_2/Mo and water. Such distributions result from a "very rapid" fragmentation process which follows an initial "slow" coarse mixing of the liquids. It was found that both explosive and non-explosive experiments produced debris with an ULLN distribution. There was no evidence of a heterogeneous distribution (a distribution composed of a mixture of two other distributions) with one distribution corresponding to slow mixing and one to fast fragmentation. These studies also showed that artificial distributions, caused by failure to recover a portion of the debris, were not being produced. In addition, a two-stage fragmentation model of the MFCI process was developed where a coarse mixing stage was followed by fine fragmentation of some of the mixture. However, it was found that a single ULLN distribution produced a better fit to the data. This distribution has the following probability density function [4],

$$f(x) = \frac{x_m}{(2\pi)^{1/2}(\ell n\sigma)x(x_m-x)} \exp\left\{-1/2\left[\frac{\ell n(x_m x/(x_m-x)) - \ell n\mu}{(\ell n\sigma)}\right]^2\right\}$$

$$0 \leqslant x \leqslant x_m \qquad (1)$$

$$= 0 \quad \text{otherwise}$$

The above distribution has three parameters, μ, σ and x_m. The parameters μ and σ give measures of location and scale, respectively. x_m is the maximum particle size which can occur. It is easily shown (by consideration of the cumulative distribution function) that the 50%, 16% and 84% points of the distribution are given by

$$x_{50} = \mu/(1 + \mu/x_m)$$

$$x_{16} = \mu/\sigma(1 + \mu/\sigma x_m) \qquad (2)$$

$$x_{84} = \sigma\mu/(1 + \sigma\mu/x_m)$$

In the limit as $x_m \rightarrow \infty$ the distribution becomes the more familiar lognormal distribution and we have

$$x_{50} = \mu$$

$$x_{16} = \mu/\sigma \qquad (3)$$

$$x_{84} = \sigma\mu$$

showing that μ and σ do indeed provide measures of location and scale. There is no simple expression for the moments of the distribution as in the case of the lognormal distribution.

3. Description of MFTF Experiments

Two series of experiments, the SUW and WUMT series, have been carried out in the Molten Fuel Test (MFTF) at AEE Winfrith. In both cases thermite generated uranium dioxide/molybdenum melt (81% UO_2, 19% Mo) at an initial temperature of 3600K in quantities of up to 24kg was used. Both series used a coolant of water.

In the SUW series the melt was released under the surface of a pool of 1.5 tonnes of water within a closed vessel. The experimental procedure and results are fully described in reference 8. For completeness the experimental conditions are given in Table 1, together with the number of steam explosions observed in each experiment and an estimate of the total work yield of the explosions.

In the WUMT series of experiments 24kg of melt was poured into a square-section mixing vessel containing water. These experiments were carried out to examine mixing; thus various combinations of melt/water mass ratio, vessel size, water depth and melt pour diameter were used in the experiments. Details of these experiments are given in reference 9 and a summary of the conditions is given in Table 2.

In both series of experiments the solidified melt debris was dried, collected from the vessel and sieved using British Standard sieves with a size range of 63 μm to 8mm. The particle size distributions plotted on log/probability scales for the SUW and WUMT series are shown in Figures 1 and 2.

For each experiment the sieved particle size distribution was fitted by an ULLN Distribution function using a non-linear least squares algorithm. The fitted distribution parameters together with an estimate of their standard error for the SUW and WUMT experiments are given in Tables 3 and 4, respectively.

3.1 Discussion of Results

The data in Tables 3 and 4 show that in general the experimental data is well fitted by the Upper Limit lognormal distribution. The most difficult parameter to fit is x_m. This is because either there is very little material at the upper size range, as is the case when an explosion has occurred so that estimation of x_m is bound to be inaccurate or there is, say, 40% of the material with a size greater than the largest sieve size, so that there is insufficient data to determine x_m. In the situations where the range of sieves covers the experimental data adequately x_m can be determined with precision as in, for example, experiments SUW07, WUMT07(R2). This is illustrated in Figure 3 which shows a comparison of the experimental and fitted distributions for experiment SUW07.

The most striking feature of the results is the wide range of experimental outcomes (violent explosion to slow boiling) which give debris which has an ULLN distribution. The data shows that in experiments where there was a steam explosion μ is typically in the range 100-1500 μm depending on the amount of material participating in the interaction. For experiments where there was no explosion μ is in the range 2-8mm. For the SUW experiments σ values are in the range 3 to 6 whereas for the WUMT experiments σ values range from 4 to 10. The estimated values of x_m show a large amount of scatter but considering only the values with a relatively small standard error gives a size range of 10-20mm. This figure is consistent with estimates of particle sizes obtained from hydrodynamic instability analysis [10].

Examination of the data does not give any clear correlation between experimental conditions and the debris distribution parameters. The results from SUW04 - SUW07 show that the debris becomes coarser as the subcooling is decreased. The effect of ambient pressure is hard to determine because although increasing the pressure appears to lead to finer debris this was a consequence of improved mixing and consequently more violent explosions [8]. The only definite (and rather obvious) result is that high steam explosion yields can be correlated with finer debris.

4. Conclusions

We have noted that in many situations in which particles are produced by fragmentation the size distribution is upper limit lognormal. The physical reasoning behind this has been discussed and numerous diverse processes leading to this distributional form have been described. In particular it has been shown that the debris from experiments where melt is quenched in a liquid coolant conform to this distribution for a range of mass scales ranging from a few milligrams to 24kgs of melt. It is noted that a wide range of experimental outcomes (ranging from a violent explosion to slow boiling) produce debris with the same functional form.

This suggests that using an upper limit lognormal distribution of particles is the appropriate choice in modelling work. It also shows that experimentalists must take great care when drawing conclusions from debris size distributions because the form of the distribution is independent of the method of debris formation, i.e. rapid fragmentation and slow chemical attack give the same functional form. Thus the shape of the debris as well as the particle size distribution needs to be reported as in reference 2.

Acknowledgement

The author would like to thank Mr. I. Cook and Dr. T.A. Dullforce of Culham Laboratory and Dr. M.J. Bird of AEE Winfrith for useful discussions on this subject.

References

[1] T.G. Theofanous and M. Saito, An assessment of class-9 (core-melt) accidents for PWR dry-containment systems. Nucl. Engrg. Des., 66, p.301-332 (1981).

[2] H. Schins and F.S. Gunnerson, Boiling and fragmentation behaviour during fuel-sodium interactions. Nucl. Engrg. Des., 92, p.221-235 (1986).

[3] A.N. Kolmogarov, Logarithmically normal distributions of fragmentory particle sizes, Dokl. Akad. Nauk. SSSR, 31, p.99-101 (1941).

[4] R.R. Irani and C.F. Callis. Particle size; Measurements, Interpretations and Applications. John Wiley and Sons, New York (1963).

[5] R.A. Mugele and M.D. Evans, Droplet size distribution in sprays. Ind. Eng. Chem., 43, p.1317-1324 (1961).

[6] D.P. Ames, R.R. Irani and C.F. Callis, The agreement between independent methods for particle size distribution measurements on finely divided powders including phosphates. J. Phys. Chem., 63, p.531-534 (1959).

[7] D.F. Fletcher, The particle size distribution of fragmented melt debris from molten fuel coolant interactions. UKAEA report AEEW-M2103 (1984).

[8] M.J. Bird, An experimental study of scaling in core melt/water interactions. Paper presented at 22nd National Heat Transfer Conference, Niagara Falls, 5-8 August, 1984.

[9] D.S.Dhuga, Private communication. (1987).

[10] D.F. Fletcher, A review of hydrodynamic instabilities and their relevance to mixing in molten fuel coolant interactions. UKAEA report AEEW-R1758 (1984).

EXPT	Release Type	Melt Mass (kg)	Cover gas volume (litres)	Absolute pressure (MPa)	Water subcooling (K)	No. of explosions	Total Yield (MJ)
01	F	24	250	0.1	78	3	0.22
02	F	24	250	0.4	87	0	0.08*
03	F	24	85	0.1	80	2	0.08
04	R	24	250	0.1	80	3	0.18
05	R	24	252	0.1	61	2	0.16
06	R	24	271	0.1	31	2	0.16
07	R	24	251	0.1	0	3	0.23
08	R	24	259	0.5	60	1	0.52
09	R	24	250	1.0	60	1	0.88
10	R	8	250	0.1	60	1	0.11
11	R	8	250	1.0	60	0	0.0
12	R	8	250	1.0	60	1	+

F = Free Release R = Restricted release using catchpot (see ref.8 for details)

* Slow pressurisation from incoherent coolant boiling

+ Not available

TABLE 1 : Test conditions for the SUW Experiments

EXPT	Water Subcooling (K)	System Pressure (MPa)	Mixing Vessel (mm)	Water Depth (mm)	Pouring Orifice (mm)	Drop Height+ (mm)	Explosion
03	80	0.1	600	500	70	0	Yes
04	0	0.1	600	485	40	15	Very small interaction
05	0	0.1	600	200	100	600	No
06	0	0.1	200	500	40	600	No
07(R2)	0	0.1	200	200	100	0	(No data)
07(R3)	0	0.1	200	200	100	0	No
09(R1)	0	0.1	600	500	100	600	(No data)
09(R2)	0	0.1	600	500	100	600	Yes

+ Due to pressurisation of the melt container there is no simple correlation between drop height and the initial velocity of the melt (see [9] for details)

TABLE 2 : Test conditions for WUMT experiments

EXPT No.	μ (μm)	Standard error	σ	Standard error	x_m (μm)	Standard error
1	754	33	3.27	0.16	26,000	22,000
2*	27,350	12,900	15.74	5.86	84,000	309,000
3	1,702	144	5.08	0.52	58,000	94,000
4	765	52	3.42	0.27	12,800	8,000
5	954	43	3.40	0.17	17,000	7,000
6	1,070	60	3.90	0.26	11,000	3,000
7	1,262	44	4.20	0.18	11,000	1,500
8	262	35	5.83	0.50	57,000	170,000
9	132	16	2.95	0.18	38,000	290,000
10	86	3	3.64	0.35	27,000	200,000
11	2,125	150	5.81	0.65	11,700	2,200
12	155	3	3.23	0.10	59,000	260,000

TABLE 3: Particle size distribution parameters for SUW experiments

* In this experiment most of the debris was recovered by breaking up a frozen melt pool on the base of the vessel so that this data should be treated with caution.

EXPT No.	(μm)	Standard error	σ	Standard error	x_m (μm)	Standard error
03	1,840	80	10.3	0.8	17,000	3,000
04	2,970	250	6.3	0.7	35,000	24,000
05	2,630	180	10.1	1.1	15,000	3,000
06	4,760	650	6.4	0.9	83,000	174,000
07(R2)	7,810	670	9.3	0.9	18,000	3,000
07(R3)	6,230	1,070	6.2	1.0	41,000	44,000
09(R1)	2,170	130	4.1	0.3	22,000	9,000
09(R2)	2,730	190	7.0	0.7	13,000	2,000

TABLE 4: Particle size distribution parameters for WUMT experiments

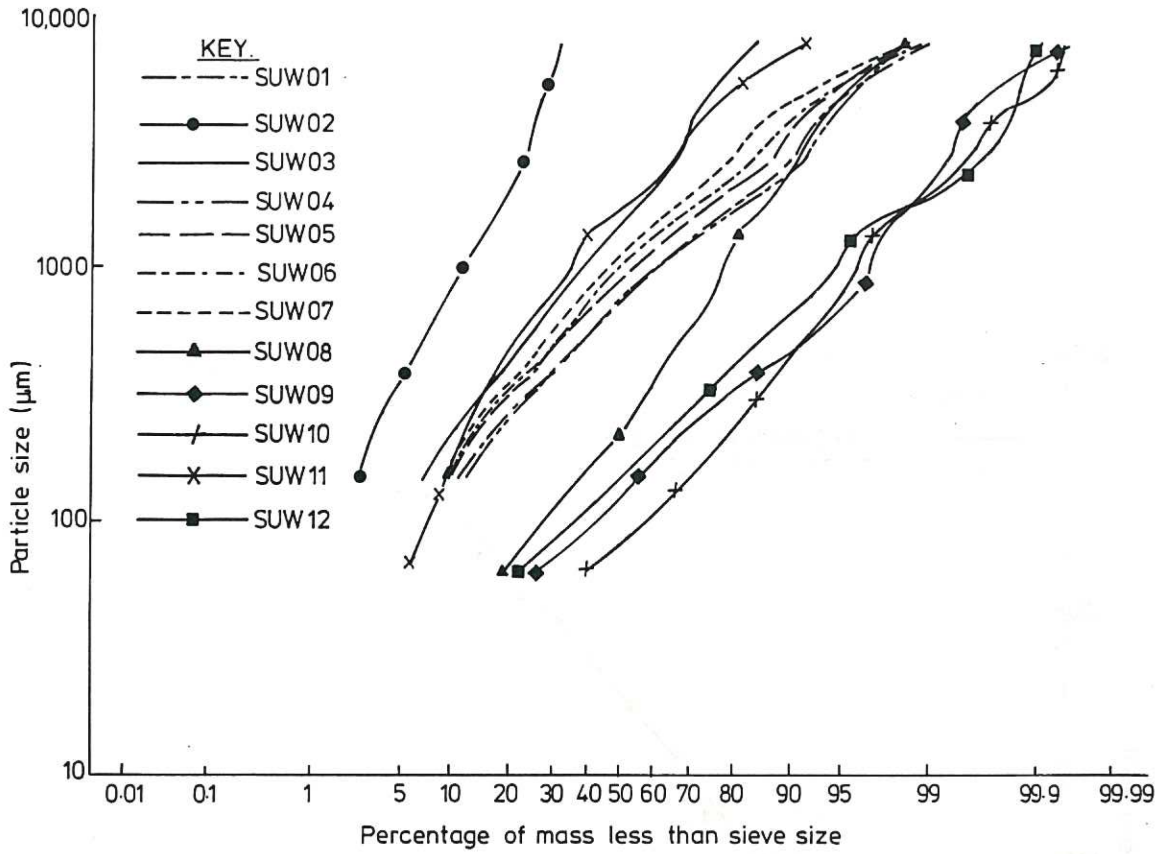

Fig.1 SUW Series Particle Size Distributions

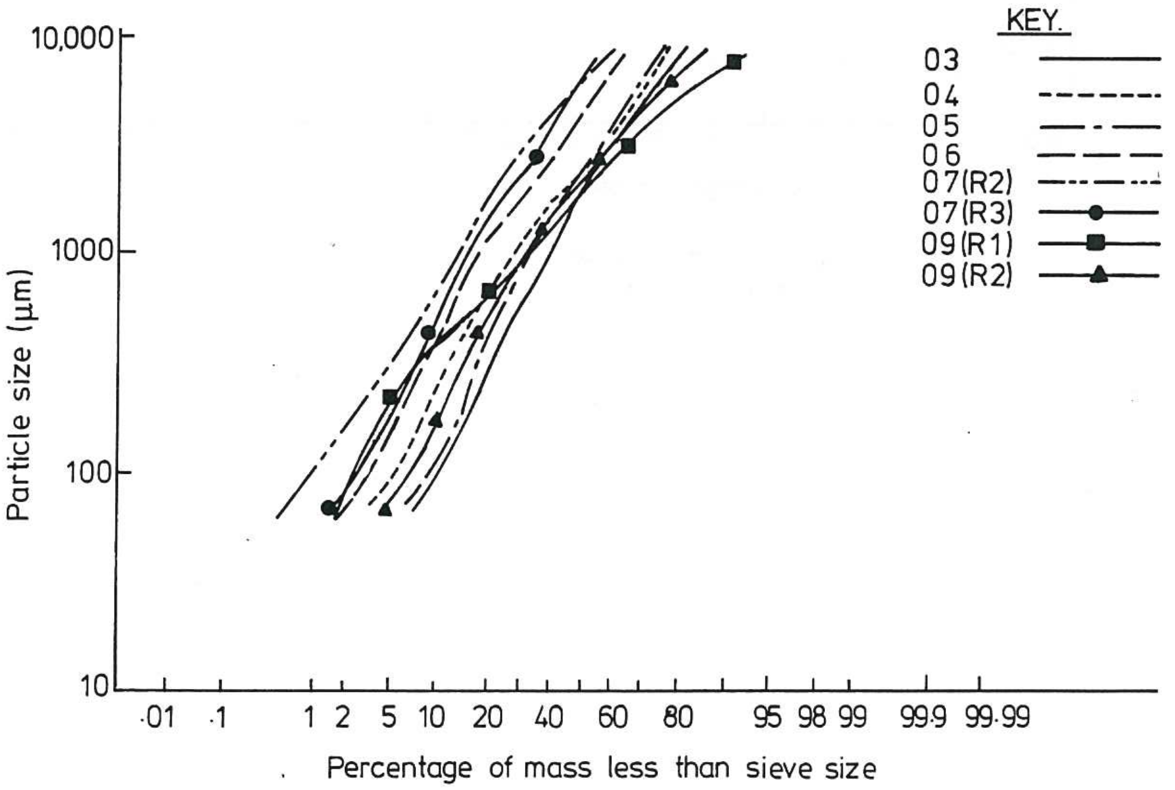

Fig.2 WUMT Series Particle Size Distributions

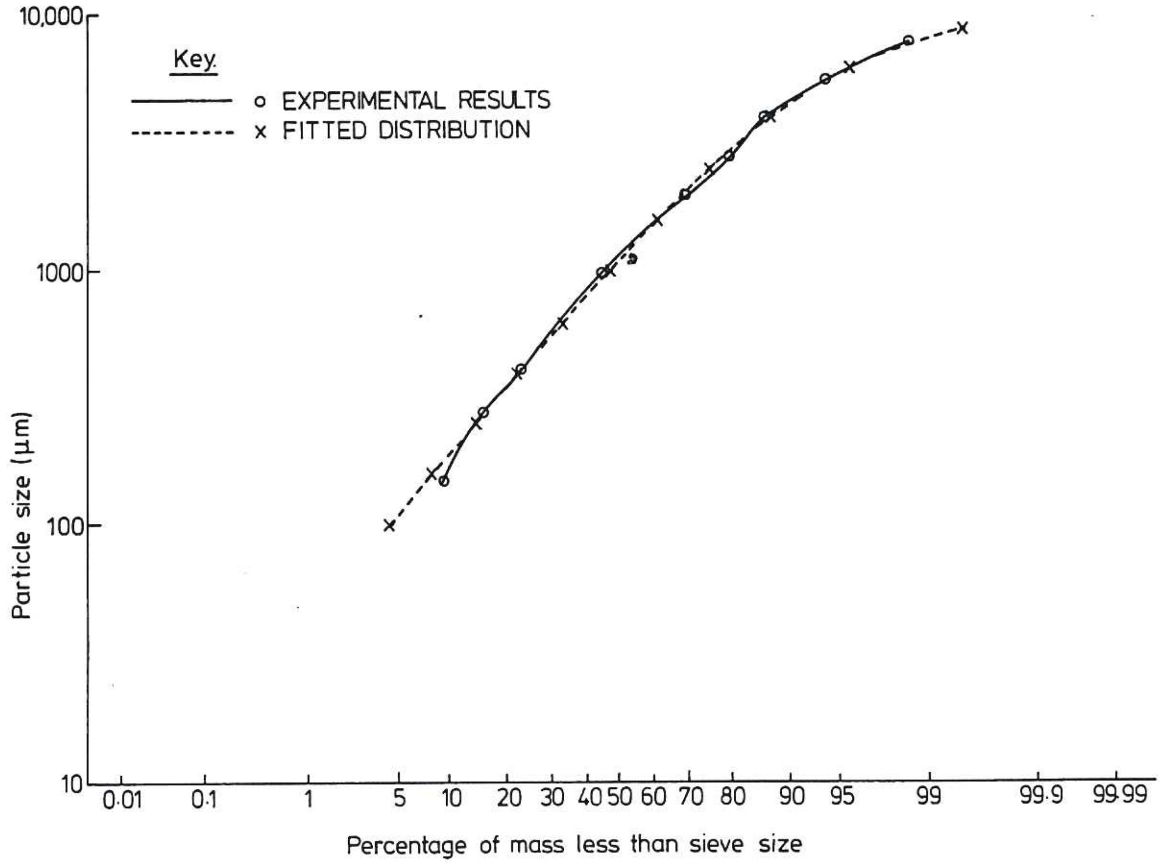

Fig.3 A Comparison of the Experimental and Fitted Distributions for Experiment SUW07.