CHARACTERIZATION OF IMPACT FRACTURE OF BRITTLE SOLID-WASTE FORMS

by

MASTER

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INTRODUCTION

The fracture of solid-waste forms resulting from mechanical impacts that could occur during in-plant handling and storage and during off-site transportation needs to be known to evaluate the increased risks of potential dispersion of radioactive wastes. Two physical properties of the waste form which may be useful to characterize the increased dispersion risks from fracture are the increase in surface area and the (mass) fraction of particles less than the respirable size (*i.e.*, $< 10\mu\text{m}$) for the waste form. An analysis and evaluation of the effects of impacts have been initiated for metal-encapsulated reference glass and ceramic waste forms. Descriptive and calculational models were developed for relating impact energy to deformation effects, both for brittle and ductile materials and for characterizing the surface area resulting from impact fracture of brittle materials¹.

This paper presents a methodology for analyzing particle size distributions obtained in impact testing of brittle waste-form materials. The methodology includes (a) a linear two-parameter lognormal correlation of the weight fraction less than any given size, (b) a mathematical function of the two lognormal parameters to determine the total surface area in terms of a dimensionless shape factor, and (c) a surface-energy constant to predict the anticipated increase of surface area from the known energy absorbed in the impact of the brittle material. Preliminary measurements were made on impacted simulated glass specimens. The analysis is used as an example of the methodology. These results are also compared with reanalyzed impact test data reported by others.

DEFORMATION OF BRITTLE MATERIALS BY MECHANICAL IMPACT

Brittle fracture of typical glass or ceramic solids under an impact results in fragmentation and formation of new surface area (ΔS_F). Fracture energy (W_F) absorbed in the brittle material is proportional to ΔS_F over a considerable range of energy input for a given specimen and mode of impact. The proportionality constant is termed¹ the fracture surface energy, γ_F . Thus,

$$W_F = \gamma_F \Delta S_F \quad (1)$$

In eq (1), W_F is generally smaller than the gross impact-energy input (W_I), due to deformation in the test equipment. Even for "bare" glass specimens impacted by hard steel surfaces, deformation of the ductile metal typically absorbs more than 50% of the gross impact energy (W_I). Variations of γ_F can be expected for different materials, sizes and shapes, and for variations of input energy concentrations in specimen volumes (V_0).

In the work on brittle fracture of Piret and associates,^{2,3,4} both single impacts and slow-compression crushing were applied to small ($\sim 1.5 \text{ cm}^3$) solid specimens (spheres and cylinders) of Pyrex glass and of prisms of natural quartz at input energy densities up to 17 J/cm^3 . Multiple impacts and slow-compression crushing were applied to particulate samples (10-14 mesh) of natural quartz. The principal result was a measurement of the net energy absorbed (W_F) in the brittle materials in producing the new surface area, as measured by a gas adsorption technique. Calorimetric methods were used to establish the net energy absorbed. Results were presented as plots of the net energy density (W_F/V_0) applied in impacting specimens vs the net fracture-surface energy, γ_F . A representative selection of the total number of data points is given in Figure 1. There is a great deal of scatter in the slow-compression data for a given test material, e.g., 10-fold variations of γ_F (4 to 80 J/m^2) for the range of values of W_F/V_0 . However, γ_F is nearly constant ($\sim 100 \text{ J/m}^2$) for impact fracture while W_F/V_0 varied over two orders of magnitude. This implies that eq (1) may be used to estimate ΔS_F for an impact input energy W_F , since γ_F is reasonably constant at most energy densities.

THE LOGNORMAL PARTICLE-SIZE DISTRIBUTION

It is shown below that the particulate solids formed in brittle fracture exhibit a particle-size distribution that can be described by the lognormal probability function. (Details as to why this is the fundamental relationship to be expected based on the physical laws will be given in a separate paper)⁵. This

Fig. 1. Results of measurements ^{2,3,4} of the fracture surface energy, γ_F , of pyrex and quartz at various specimen net impact energy densities for compression tests [CT], single impact tests [SIT] and multiple impact tests [MIT].

function has two parameters obtainable from graphical analysis, namely the geometric mean diameter D_g and the geometric standard deviation σ_g . D_g is the diameter at the 50% mass fraction, and σ_g is the ratio of the diameters plotted at 50% and 16% mass fractions. The ratio (S_F/V_0) of total particulate surface area to solid volume, can be expressed⁶ as a particulate "equation of state" in terms of the lognormal parameters as

$$\frac{S_F}{V_0} = \frac{\alpha}{D_g} \sigma_g^{0.5 \ln \sigma_g} \quad (2)$$

where α is a dimensionless shape factor. The minimum value of α is 6 (for spheres and cubes). For brittle fragments of other shapes, α is typically 2 to 3 times higher. The total particulate surface area S_F is related to the original specimen surface area S_0 and to the surface area formed in fracture, ΔS_F , as:

$$\Delta S_F = S_F - S_0. \quad (3)$$

The relations expressed by the three above equations provide quantitative parameters which can serve as a basis for systematic test programs for characterizing the results of impact on various composite bodies of brittle and ductile materials, such as canistered glass waste forms or metal encapsulated composites.

APPLICATION OF THE ENERGY AND SIZE RELATIONSHIPS TO IMPACT TESTING

Two 5.3-g samples consisting of 19 Pyrex spheres (6.2 mm) were each impacted by a drop weight device at two energy densities 6.9×10^6 and 3.3×10^6 J/m³, based on gross impact energy input. The net energy input was not measured. Size distributions were measured by sieve analyses and are labeled ANL-1 and ANL-2 in Table 1. The fracture surface area was also measured for one test (ANL-2) by (Kr) gas BET adsorption. The cumulative lognormal plot of these particle size data are given in Fig. 2. The linearity of the plots is important for estimating the cumulative mass fraction of respirable sizes (<10 μ m). The lognormal parameters from the graphical analysis and the calculated fracture-surface energies are also shown in Table 1. The fracture surface area for test ANL-1 was calculated from the lognormal surface/volume ratio (S_F/V_0 in Table 1) with the value of the shape factor $\alpha = 12$ as determined from test ANL-1.

For comparison, also reported in Table 1 and Fig. 2, are data selected from the reported literature on the impact fracture of brittle materials representative of solid waste forms. Impact tests on two materials at two different energy densities made at the Savannah River Laboratory (SRL)⁷: were (1) a simulated borosilicate waste-glass as a cylindrical specimen 0.50 inch diameter and 1.0 inch long and (2) a concrete/cylinder consisting of "neat" high-alumina cement 1.0 inch diameter and 0.50 inch long. Two tests made at Battelle Pacific Northwest Laboratory (PNL)⁸ used a reference commercial soda-lime silica glass cylinder 0.44 inch diameter and 0.50 inch long. Engineering studies of impact effects on fuel capsules (for aerospace applications) were made at Monsanto's Mound Laboratory (ML)⁹; two impact tests of PuO₂ microspheres sealed into a stainless steel (6 1/2" long x 7/8" O.D. with 3/8" walls) capsule were reanalyzed. Only data from the glass specimens are reported in Table 1.

Table 1. Selected Test Data on Impact Fracture of Brittle Specimens

KEY	⊙	⊙	⊠	⊠	◇	◇
INDEX	ANL-1	ANL-2	SRL-1	SRL-2	PNL-1	PNL-2
MATERIAL	Pyrex Beads	Pyrex Beads	B-Glass Cyl	B-Glass Cyl	SL Glass Cyl	SL Glass Cyl
VOLUME V_0 (cm ³)	2.38	2.38	3.33	3.33	1.25	1.25
SURFACE S_0 (cm ²)	23	23	12.7	12.7	12.8	12.8
SURFACE S_F (cm ²)	424 (calc'd)	1000 (meas'd)	354 (calc'd)	1320 (calc'd)	1540 (meas'd)	2870 (meas'd)
ENERGY $\frac{H_I}{V_0}$ ($\frac{J}{m^3}$)	3.3×10^6	6.9×10^6	5.9×10^6	2.4×10^7	2.2×10^7	8.9×10^7
Lognormal parameters:						
$D_g(\mu m)$	2100	930	7000	750	470	225
σ_g	4.4	4.6	6.7	3.8	3.8	3.8
Wt % $D \leq 10 \mu m$	0.02	0.32	0.03	0.06	0.20	0.90
$\frac{S_F}{V_0}$ ($\frac{l}{m}$)	1.46×10^3	3.44×10^3	8.72×10^2	3.25×10^3	5.19×10^3	1.08×10^4
γ_F ($\frac{J}{m^2}$)	1.8×10^2	1.6×10^2	5.6×10^2	6.0×10^2	1.8×10^2	3.9×10^2

Fig. 2. The cumulative lognormal plot of measured particle size distributions for various impact tests on glass materials.

However, all particle size data, when replotted lognormally as in Fig. 2, formed straight lines and all values of σ_g (from graphical analysis) were in the range shown for the glass specimens.

The analysis of the above data shows that for a given material, σ_g is nearly constant over a range of energy densities.

The D_g varies inversely with the energy density. The surface area as determined directly by gas adsorption or as calculated from the lognormal parameters and the shape factor $\alpha = 12$ allows calculation of a fracture-surface energy γ_F (J/m^2) if the net energy is known. For impacts, γ_F appears to be nearly constant for a given material, and to increase at higher energy densities (W_F/V_O). The energies reported here are the total input energies; the net energy absorbed in brittle fracture are expected to be lower by a factor of about two for "bare" specimens and a factor of more than four for canistered specimens. The considerable regularities in lognormal relationships of these tests of different specimens under quite varied test conditions indicate promise as a basis of further systematic tests and analyses.

Three different glass compositions of small specimens were impacted⁸ at four different energies. Particle sizes were measured by sieving and the original results were presented in nonlinear log-log plots. Fracture surfaces were not reported. To test the methodology on different glass compositions, the particle size data were replotted on lognormal graphs to determine the two lognormal parameters and the surface area. These lognormal plots are shown in Fig. 3, and the lognormal parameters, calculated surface areas and fracture-surface energies are shown in Table 2. From reported data⁸ on surface areas of typical particle size distributions of soda-lime-silica glass, a mean shape factor of $\alpha = 21 \pm 3$ was calculated. This value of α was used to calculate surface areas from the lognormal S_F/V_O ratio given in eq (2). These parameters show the comparative fracture properties of the materials. The constancy of the values of σ_g is striking, as shown by the similar slope in Fig. 3.

To obtain a range of impact energies, multiple impacts on a single specimen have been used at SRL and PNL. For example, tests of the same type of specimen were made with the same total energy-input density, but with that total energy applied in either 1, 2, 4, and 8 blows⁸. The size distributions have been plotted lognormally in Fig. 4. From the lognormal parameters, the highest energy density (1 blow) showed the smallest σ_g , the largest γ_F , and the smallest S_F/V_O . The principal difference was the respirable fraction ($< 10 \mu m$ diam.) which was 1.3% for 1 blow and 6.6% for 8 blows. This effect is consistent with other brittle-fracture data.

Fig. 3. Lognormal plots of data reported⁸ for three types of impacted glass compositions at four energy densities.

Table 2. Lognormal Parameters of Impact Fracture of Three Different Glass Compositions

Glass Type	W/V_0 (J/m ³)	D_g (μm)	σ_g	Wt% $D < 10 \mu m$	S^a $\frac{S}{V}$	Calc YF (J/m ²)
Soda-lime-silica Glass	1.70×10^8	192	3.7	1.2	3.75×10^5	4.5×10^2
	8.9×10^7	220	3.7	0.8	2.47×10^5	3.6×10^2
	4.6×10^7	370	4.1	0.6	1.65×10^5	2.8×10^2
	2.2×10^7	490	4.4	0.4	1.19×10^5	1.8×10^2
Frit	1.7×10^8	290	4.3	1.0	2.1×10^5	8.1×10^2
	8.9×10^7	490	4.4	0.4	1.3×10^5	6.8×10^2
	4.6×10^7	530	4.4	0.4	1.2×10^5	3.8×10^2
	2.2×10^7	720	4.2	0.2	8.3×10^4	2.7×10^2
Fused Silica (vitreous)	1.7×10^8	135	4.1	3.2	4.2×10^5	4.0×10^2
	8.9×10^7	150	4.0	2.8	3.7×10^5	2.4×10^2
	4.6×10^7	220	4.0	1.4	2.5×10^5	1.8×10^2
	2.2×10^7	300	4.0	0.6	1.9×10^5	1.2×10^2

Fig. 4. Lognormal plot of data reported⁸ for impacted glass specimens receiving the same total energy but with a different number of impacts.

CONCLUSIONS

Data obtained from impact fracture of brittle materials can be correlated with a linear lognormal particle size distribution and with a nearly constant brittle-fracture surface-energy factor. This methodology allows estimation of total surface area of particles from impact fracture. Reliable extrapolations of particle size distributions to any lognormal size range seems achievable. Confirmation of this methodology are subjects of current study.

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