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A modified Weibull treatment for the analysis of strength-test data from non-identical brittle specimens

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Powder compacts (e.g., pharmaceutical tablets) manufactured on commerically available machines are not strictly identical but show inevitable variability in their weights, thicknesses and compaction pressures. Consequently, the variability in fracture-stress data obtained from such brittle specimens is greater than that due to the inherent strength variability of the material itself. A modified Weibull analysis has been developed so that a more accurate estimate of the inherent variability of the mechanical strength of the material can be derived from test data obtained from commercially produced compacts; its application is illustrated.

Nomenclature

D	diameter
$f(\rho v)$	relative frequency of occurrence of specimens with density ρ and volume v
F	minimization function
i	ascending rank number of a fracture stress
m	Weibull modulus
N_{tot}	number of specimens in a batch
$N(\rho v)$	number of specimens with densities in the range ρ to $\rho + d\rho$ and volumes in the range v to $v + dv$
P_f	failure probability
p_u	upper punch compaction pressure
t	thickness
v	volume
w	weight
W_f	fracture load
ρ	density
σ_f	fracture stress
$\bar{\sigma}_f$	mean fracture stress of a batch
$\bar{\sigma}_f(\rho v)$	mean fracture stress of specimens with density ρ and volume v
σ_0	scale parameter or normalizing factor
σ_u	location parameter or threshold stress

1. Introduction

It is well established that nominally identical brittle specimens have variable fracture stresses both in simple uniaxial tension [1] and in more complex test modes. This observed variability is attributed to the different severity of the strength-governing flaws [2] in the different specimens of a batch and is directly associated with the brittleness (i.e., the lack of ductility) of the material. The Weibull distribution function [3] has been widely used in attempts to characterize the fracture-stress variability.

Powder compacts behave as brittle materials in mechanical strength tests (e.g., the diametrical compression test [4]) in that a range of fracture stresses are obtained from nominally identical

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specimens, and previous work [5] has indicated that the fracture stresses of such specimens can be satisfactorily represented by a Weibull distribution.

A recent study [6] has examined the strength characteristics of powder compacts produced by single-ended compaction in commercially available production machines. These specimens cannot be considered nominally identical since they show significant variability in such properties as weight, thickness and density, associated with random variations in die-fill weight. The characterization of the fracture stresses of such specimens in terms of the unmodified Weibull distribution will give inaccurate and misleading Weibull parameter estimates [7] and consequently, if more reliable estimates of these parameters are required, a modified Weibull distribution function must be developed in which allowance is made for variations in the above properties. Such a modification is presented in this paper and its application in the analysis of strength-test results for a large batch of aspirin tablets is described.

2. The Weibull distribution

The distribution function atrributed to Weibull [3] refers to a large batch of nominally identical specimens and relates the cumulative failure probability, P_f, to the fracture stress, σ_f, by the use of three independent parameters in the form:

$$P_f = 1 - \exp\left[-\frac{\sigma_f - \sigma_u}{\sigma_0}\right]^m. \quad (1)$$

In this expression, m, the "Weibull modulus", is a reciprocal measure of fracture stress variability about the mean value, functionally related to the standard deviation of the distribution. σ_0 is the "scale parameter" or "normalizing factor"; it does not correspond directly to any readily determined physical quantity but it can be shown [8] that it is related to the arithmetic mean of the fracture-stress distribution ($\bar{\sigma}_f$) by the equation:

$$\bar{\sigma}_f = \sigma_0 \left(\frac{1}{m}!\right) \quad (2)$$

where $[(1/m)!]$ is the "gamma" function of $[(1/m)+1]$ [9]. σ_u is the "location parameter" or "threshold stress", i.e., the minimum stress for which fracture can occur. For stresses smaller than σ_u the failure probability is zero. It is usual in Weibull model-based work to assume that σ_u is zero. With this assumption, Equations 1 and 2 reduce to the following equation:

$$P_f = 1 - \exp\left[-\left(\frac{1}{m}!\right)^m \left(\frac{\sigma_f}{\bar{\sigma}_f}\right)^m\right]. \quad (3)$$

This is the standardized form of the Weibull distribution function, relating failure probability to fracture stress and characterized by the two parameters m and $\bar{\sigma}_f$. It is applicable in cases where the applied stress is uniaxial and uniform. For complex stress systems (e.g. the diametral compression test) the form of Equation 3 is unchanged if the "fracture stress", σ_f, is taken to be proportional to the fracture load, W_f. In the results presented here the relationship

$$\sigma_f = \frac{2W_f}{\pi Dt} \quad (4)$$

has been used throughout and the analysis has been based on Equation 3.

3. Estimation of distribution parameters from fracture-stress data

Clearly there is no difficulty in estimating the mean fracture stress from experimental data, but the estimation of the Weibull modulus is not so straightforward. Several procedures are available for this purpose [10]; the three most widely used are:

(i) a linearization technique in which m is obtained as the slope of the $\ln\ln(1/(1-P_f))$ versus $\ln\sigma_f$ linear plot;

(ii) direct curve fitting using the "least squares" method;

(iii) direct curve fitting using the "maximum likelihood" method.

A number of publications (e.g., [10–12]) have compared these and other methods of estimating m and there is no strong evidence that one method is significantly better than the others. In the work presented here a computer program incorporating the "least squares" method was utilized which estimated m in the following way:

(i) the values of fracture stress, σ_f, for the specimens under consideration are "ranked" in order of increasing magnitude;

(ii) the failure probability $[P_f(i)]$ corresponding to the ith fracture stress level is obtained from the relationship

$$P_f(i) = \frac{i}{N_{tot} + 1}, \quad (5)$$

where N_{tot} is the number of specimens in the batch;

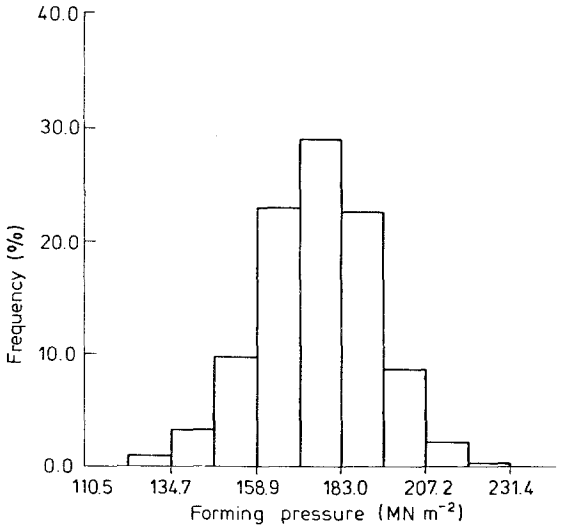

Figure 1 Histogram of the forming pressure of 1005 Aspirin tablets.

(iii) using an assumed value of m the failure probability $P_f^*(i)$ for each nominal fracture stress level is obtained from Equation 3;

(iv) the function

$$F = \sum_{i=1}^{i=n} (P_f^*(i) - P_f(i))^2 \qquad (6)$$

is evaluated;

(v) using a minimization routine the value of m in Equation 3 is adjusted until the function F has a minimum value. The corresponding value of m is the "least squares fit" value of the Weibull modulus.

4. Experimental work

A batch of 1005 powder compacts, of 10 mm diameter, was prepared by single-ended compaction using a commercially available punch and die machine† from a $-500+425\,\mu m$ size fraction of acetylsalicylic acid (Monsanto Crystalline Aspirin – 7016). A load cell containing strain gauges had been fitted behind the upper punch of the compaction machine so that a quantitative measure of the force applied in compacting each specimen could be obtained.

The compacting force for each tablet was determined to a precision of 1% from an ultraviolet galvanometer. There were no detectable differences in the diameters of a random sample of the tablets as measured to ±0.01 mm with a micrometer. Each tablet was weighed to ± 0.0001 g on an electrical balance and the thickness was measured to ±0.01 mm using a micrometer. After manufacture the tablets were stored in airtight containers for a period of 2 weeks before their tensile fracture stresses were determined by means of the diametral compression test [4], using a test machine‡ designed specifically for this purpose.

5. Experimental results

Histrograms of the distributions of forming pressure (p_u), weight (w), thickness (t) and tensile fracture stress (σ_f), are shown in Figs 1 to 4, and the corresponding mean values, standard deviations and coefficients of variation and the derived volumes (v) and densities (ρ) are given in Table I.

6. Analysis

It is clear from these results that the properties of the tablets in this large batch were not strictly uniform and that the tablets could not properly be described as "nominally identical" in the intended sense. (These variations in properties arise from the nature of the manufacturing process in which the powder is fed into the punch-die cavity under its own weight via the "feed shoe" of the tabletting machine. Inevitable variations in the flow behaviour of the crystalline aspirin result in variations in die-fill and consequently in tablet weight, etc.).

Because of the variations in density and volume of the tablets within the batch, the observed strength variability cannot be entirely attributed to the intrinsic strength variability of the tablet material. It is well established [8] that volume variations alone, for example, *ceteris paribus*, will give rise to variations in the tensile fracture stress. It is to be anticipated, therefore, that the value of the Weibull modulus derived from the test data without regard to the variations will be systematically incorrect.

In order to demonstrate this effect and to indicate the magnitude of the error involved, Weibull modulus values were determined for (i) the entire batch (with no allowance for differences in volume and density), and (ii) 31 selected tablets with almost identical properties (the compaction pressures could not be differentiated and the coefficients of variation in weight and thickness were 0.22% and 0.13%, respectively). The modulus

†Manesty Machines Ltd. Liverpool, UK – Model F3.
‡Engineering System (Nottingham) Ltd, UK – Model CT40.

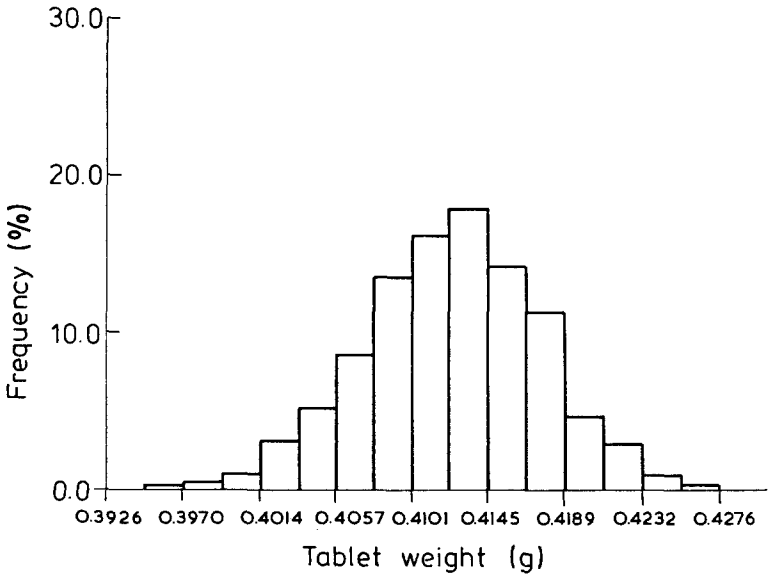

Figure 2 Histogram of the weight of 1005 Aspirin tablets.

values obtained were (i) 12.23 ± 0.27, and (ii) 15.54 ± 1.97. In spite of the greater probable error in the latter value, due to the much smaller sample size, these figures do suggest that non-uniformity of specimen properties may significantly affect the Weibull modulus estimate. Thus, if a more accurate estimate of this quantity is to be obtained then a modified analysis is required which must allow for factors, other than the inherent material strength variability, which may influence the fracture stress of specimens.

In this study these factors have been represented by volume and density. It is known that fracture stress is dependent on specimen volume [5] and a relationship between the mean fracture stresses of two batches of similar brittle specimens of different volume has been proposed. The weight, thickness and forming pressure of tablets produced in the simple fixed-punch displacement machine used in this work are interrelated. A change in either one of these quantities will give rise to a tablet of different density. Therefore, changes in tablet density have been used in the modified analysis presented here to reflect the random changes that occur in weight, thickness and forming pressure.

A modified Weibull distribution function, in which the volume dependence and density dependence of fracture stress have been incorporated,

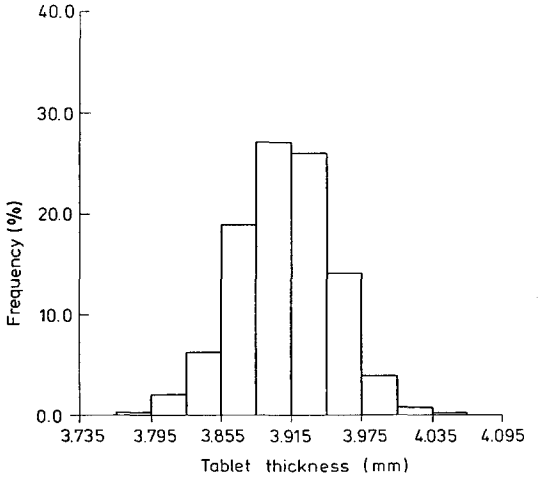

Figure 3 Histogram of the thickness of 1005 Aspirin tablets.

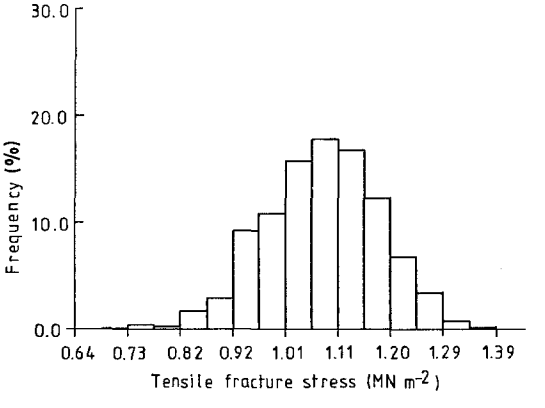

Figure 4 Histogram of the tensile fracture stress of 1005 Aspirin tablets.

TABLE I

	Mean	S.D.	Coefficient of variation
Compaction pressure (p_u)	176.3 MN m^{-2}	16.6 MN m^{-2}	9.41%
Weight (w)	0.4123 g	0.0050 g	1.22%
Thickness (t)	3.91 mm	0.04 mm	1.04%
Fracture stress (σ_f)	1.077 MN m^{-2}	0.103 MN m^{-2}	9.53%
Volume (v)	309 mm^3	3.2 mm^3	1.04%
Density (ρ)	1.332 Mg m^{-3}	0.0055 Mg m^{-3}	0.41%

has been published [13] and is derived in the Appendix. The modified function is:

$$P_f = \int_0^\infty \int_0^\infty f(\rho v) \left\{ 1 - \exp\left[-\left(\frac{1}{m}!\right)^m \times \left(\frac{\sigma_f}{\bar{\sigma}_f(\rho v)} \right)^m \right] \right\} dv\, d\rho, \quad (7)$$

in which the failure probability, P_f, of specimens varying in volume and density is related to the associated fracture stress, σ_f. The parameters $\bar{\sigma}_f(\rho v)$ and $f(\rho v)$, respectively, are the mean fracture stress and relative frequency of occurrence of specimens with density ρ and volume v, m is the Weibull modulus and $[(1/m)!]$ is the "gamma" function of $[(1/m) + 1]$.

The integral in Equation 7 can be evaluated either analytically or numerically. For analytical integration the distribution function $f(\rho v)$ has to be expressed mathematically. It is known that the distribution function of tablet density is complex, with significant skewness and kurtosis, and since it would be very difficult to represent this distribution function mathematically, and to integrate the resulting expression, a numerical evaluation of Equation 7 has been adopted.

The distribution curve of a single variable can be represented graphically by a frequency distribution. In the present case, a two-variable histogram is required to depict a distribution "surface" defined by the two variables, volume and density. Sections through such a surface are shown in Fig. 5. The number of specimens, $N(\rho v)$ with densities in the range ρ to $\rho + \Delta\rho$ and volumes in the range v to $v + \Delta v$ is given by:

$$N(\rho v) = f(\rho v)\, \Delta\rho\, \Delta v N_{tot},$$

i.e.

$$\frac{N(\rho v)}{N_{tot}} = f(\rho v)\, \Delta\rho\, \Delta v, \quad (8)$$

where N_{tot} is the total number of specimens.

Equation 7 can be evaluated numerically by constructing a two-variable frequency distribution

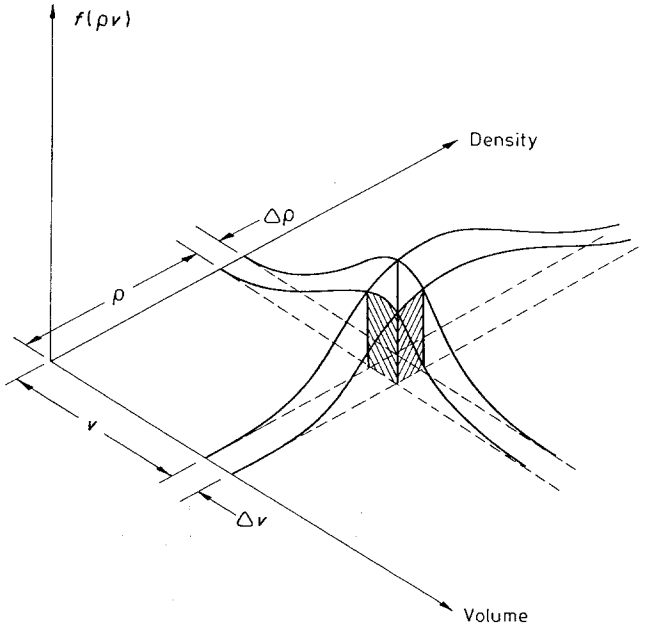

Figure 5 Sections through a bivariate frequency distribution of volume and density.

for the test data. This is done by dividing the range of densities into k equal intervals of $\Delta\rho$ and the range of volumes into k equal intervals of Δv, giving k^2 frequency distribution "blocks". If the central values of each density and volume class interval are $\rho_1, \rho_2, \rho_3, \ldots, \rho_h, \ldots, \rho_k$, and $v_1, v_2, v_3, \ldots, v_j, \ldots, v_k$, respectively, and there are N_{hj} tablets in each "block" with central values $\rho_h v_j$ then:

$$\frac{N_{hj}}{N_{\text{tot}}} = f(\rho_h v_j)\,\Delta\rho\,\Delta v$$

and Equation 7 becomes

$$P_f = \sum_{j=1}^{j=k}\sum_{h=1}^{h=k}\frac{N_{hj}}{N_{\text{tot}}}\left\{1-\exp\left[-\left(\frac{1}{m}!\right)^m \times\left(\frac{\sigma_f}{\bar{\sigma}_f(\rho_h v_j)}\right)^m\right]\right\} \quad (9)$$

where $\bar{\sigma}_f(\rho_h v_j)$ is the mean fracture stress of the specimens within the frequency distribution "block" with central values $\rho_h v_j$.

7. Application of the modified analysis

The "least squares fit" value of the Weibull modulus, when corrected for variability in tablet density and tablet volume (i.e., the "corrected" m value), can be calculated in a similar manner to that given previously for the unmodified Weibull modulus. The procedure is given below.

(i) The test data are ranked with respect to fracture stress, and the failure probability from mean ranking, $P_f(i)$, for each of the N_{tot} stress levels is obtained as in Equation 5.

(ii) The data are sorted into density and volume frequency distribution "blocks".

(iii) The value of

$$\frac{N_{hj}}{N_{\text{tot}}}\left\{1-\exp\left[-\left(\frac{1}{m}!\right)^m\left(\frac{\sigma_f}{\bar{\sigma}_f(\rho_h v_j)}\right)^m\right]\right\}$$

is evaluated for each histogram "block" using an assumed value of m.

(iv) The failure probability, $P_f^*(i)$, given by Equation 9 is obtained by summing the values of the above expression for each bivariate frequency distribution "block".

(v) The function

$$F = \sum_{1}^{n}(P_f^*(i) - P_f(i))^2$$

is evaluated.

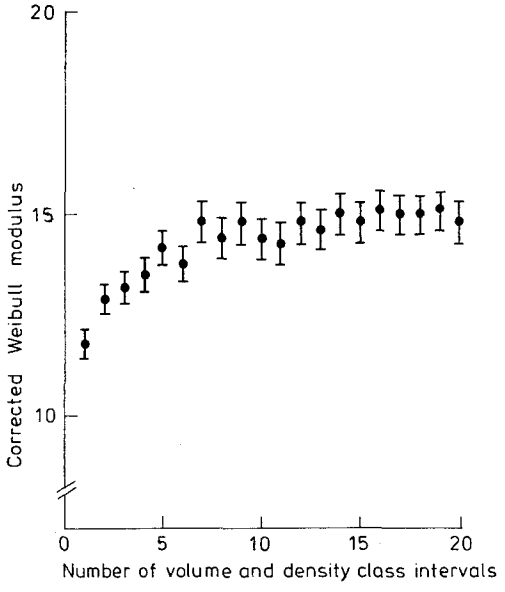

Figure 6 The dependence of the corrected Weibull modulus on the number of class intervals from a sample of 500 taken from the 1005 tablets.

(vi) An iterative procedure is employed and the value of m changed until the function F has been minimized. The corresponding m value is the "least squares fit" value derived using the modified Weibull distribution function.

The modified analysis has been applied to the batch of 1005 aspirin tablets. In order to examine the adequacy of the "block" representation of the bivariate frequency distribution, the number of blocks used in the analysis was systematically varied by splitting the density and volume ranges into 1, 2, 3, ..., 20 intervals, giving 1^2, 2^2, 3^2, ..., 20^2 blocks. Further, in order to assess the importance of sample size, corrected modulus values were determined for groups of 30, 50, 100, 150, 200 and 500 selected in a systematic manner from the whole batch. A sample of the results of this preliminary analysis is shown in Fig. 6 in which the corrected Weibull modulus, with standard error bands, is plotted against the number of class intervals used in the sub-division of the density and volume ranges for a sample of 500 tablets. It was suggested by this work that there was an optimum number of bivariate histogram blocks for this modified analysis which depends on the sample size. Fig. 7 shows the correct Weibull modulus values plotted against the average number of data per histogram block for the different sample sizes studied. With an average of less than

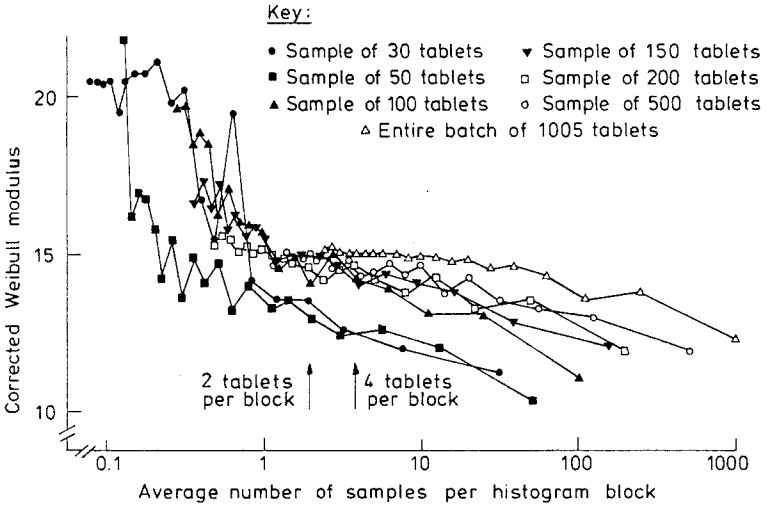

Figure 7 The corrected Weibull modulus values plotted against the average number of tablets per block for different sample sizes.

one data point per block, the derived modulus values vary erratically. With 2 to 4 data per block the corrected modulus values are not sensitive to this quantity and are not markedly dependent on sample size for samples greater than 100. It appears that samples less than 100 in size may not be adequate to represent the bivariate frequency distribution. As the number of data points per block increases the corrected modulus falls from 15 (approximately) to 12 (approximately). (The 1 x 1 block sub-division is equivalent to the unmodified analysis for which an m value of 12.23 was obtained.) It was concluded that for a reliable estimate of the corrected Weibull modulus there should be 2–4 data points on average per histogram block and a minimum sample size of 100 should be used.

The "best" corrected Weibull modulus value obtained using the entire batch was 15.16 ± 0.34. This value agreed well with the value of 15.54 ± 1.97 obtained from 31 specimens with almost identical properties and is considerably larger than the value of 12.23 obtained from the unmodified Weibull analysis. The corrected Weibull modulus of 15.16 is to be regarded as a satisfactory measure of the intrinsic material strength variability derived from these non-identical specimens.

Appendix. Derivation of the modified Weibull distribution function

A modified Weibull distributed function relating failure probability to stress is developed for specimens which exhibit variability in their densities and their volumes.

Let $f(\rho v)$ be the relative frequency of specimens with density ρ and volume v. If N_{tot} tablets are tested up to a stress, σ_f, then the number in the density and volume ranges ρ to $\rho + \Delta\rho$ and v to $v + \Delta v$, which fail, is given by:

$$\text{number to fail} = f(\rho v)\,\Delta\rho\,\Delta v\,N_{tot}P_f(\sigma_f)$$

where $P_f(\sigma_f)$ is the failure probability of nominally identical specimens at stress σ_f.

Therefore, the total number of tablets to fail through the entire density range with volume v to $v + \Delta v$

$$= \int_0^\infty f(\rho v)P_f(\sigma_f)N_{tot}\,\Delta v\,d\rho,$$

and the total fail through the entire volume range

$$= \int_0^\infty \int_0^\infty f(\rho v)P_f(\sigma_f)N_{tot}\,dv\,d\rho,$$

Therefore,

$$\text{failure probability} = \frac{\text{number of specimens to fail}}{\text{total number of specimens}}$$

$$= \int_0^\infty \int_0^\infty f(\rho v)\,P_f(\sigma_f)\,dv\,d\rho.$$

But,

$$P_f(\sigma_f) = 1 - \exp\left[-\left(\frac{1}{m}!\right)^m \left(\frac{\sigma_f}{\bar{\sigma}_f(\rho v)}\right)^m\right].$$

Therefore, the failure probability P_f at a stress of σ_f for these non-identical specimens is given by:

$$P_f = \int_0^\infty \int_0^\infty f(\rho v) \times \left\{1 - \exp\left[-\left(\frac{1}{m}!\right)^m \left(\frac{\sigma_f}{\bar{\sigma}_f(\rho v)}\right)^m\right]\right\} dv\,d\rho,$$

where $\bar{\sigma}_f(\rho v)$ is the mean fracture stress of specimens with density ρ and volume v, and $[(1/m)!]$ is the "gamma" function of $[(1/m) + 1]$.

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