Using a Population Balance Model Coupled with Structural Effects to Aid Product Design of Bran Cereal

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Abstract

Particle size degradation is a mechanistic based process that is a function of the structure of individual agglomerates in the system. Degradation can be caused by stress and strain events or impact events. Causes of material breakage include fracture of agglomerates to smaller particles and knocking off corners of large particles in abrasion events. To mitigate degradation of a given product, one must understand the relationship between structure and the causes of degradation. Industry often uses single point indices tests to estimate particle size degradation in typical processes. This provides an accurate estimate only if the actual cause of degradation in the process matches that measured with the particular tester. The population balance modeling technique provides a powerful tool to determine the root cause of size degradation of a given material and provide some guidance on how the material should be changed to reduce degradation. This paper looks at size degradation from both a simple index approach and a population balance model coupled with a particle structural analysis. Each analysis provides information to the engineer concerning the behavior of a given product in a prescribed process. The population balance model provides additional information regarding the cause of particle breakage. This paper looks at the advantages and disadvantages of both methods using bran cereal as an example material.

Key Words

Size degradation, Abrasion, Agglomerate structure, Fracture, Bulk Material

Introduction

The use of nano-particles in bulk solid materials has led to enhanced chemical and structural ability of these materials. There is a movement in industry to create large size particles (agglomerates) with a high degree of porosity and large surface areas where nano-particles on the surface perform various functions. Herein lies the dilemma. Large particles are preferred due to the lack of cohesive flow problems. However, large particles have low surface area and, hence, limited chemical functionality. One solution to this particle design process quandary is to use an agglomeration process that creates a large porous particle from a series of small nano-particles. Unfortunately, these engineered materials are generally both highly porous and friable, resulting in significant degradation during use. Whatever the reason, degradation of particles is a problem in today's production facilities. Degradation produces fines that limit the life of catalyst in fluid bed operations. Size degradation causes fines production in pharmaceutical products that

subsequently segregate, resulting in quality issues. Particle size degradation causes consumer acceptance issues with many consumer products from food to bath products. Degradation issues also lead to dust generation problems, environmental discharge, and safety concerns.

Thus, measuring and understanding degradation is critical to engineers and materials scientists designing processes and products that minimize these effects. There are several key issues to developing a robust method to mitigate size degradation effects. Degradation is caused by different external influences. Placing material in a condition where it is exposed to large stresses and high strains causes reduction in particle size due to particle breakage under the stress/strain behavior. Impact can also result in particle breakage.

It is critical to match the degradation measured by a particular method to the type of degradation present in the unique plant production facility. Failure to do this results in situations where the degradation test technique predicts a certain amount and form of degradation which will not actually occur in the process. If impact during processing dominates the process flow behavior, then degradation due to impact behavior is the critical property to measure, in order to provide predictive size reduction estimates. Using a degradation tester that causes size reduction due to stress and strain behavior may give erroneous results when applied to processes which are impact dominated. Although these degradation principles are common to many industries, we have chosen to examine the bran cereal as an example product. Two types of bran particles will be considered in this paper. We performed degradation analysis on bran agglomerates consisting of particles measuring about 4500 microns with an aspect ratio of about 1:1. These will be referred to as bran pearls in this paper. We also performed degradation analysis on an extruded bran product with a diameter of about 2200 microns and a length around 14,000 microns. These will be referred to as bran fibers in this paper.

Simple Process Degradation Index Test

One method used to quantify the degradation occurring with a particular material is to impose a constant stress/strain condition or a constant impact condition on a material and monitor the shift in particle size distribution due to the applied stimulus. There are many such tests in industry used to accomplish this process. Consider, for example, the work of Barletta, et al [1]. Here the critical particle size was chosen and material was subjected to impacts (tapping). The shift in particle size was measured. They found a power law response to this type of degradation when used with powdered milk. We also subjected bran pearls to repeated impacts using a different tester and monitored the shift in particle size. For example, Figure 1 shows a typical shift in the cumulative particle size as bran pearls are subjected to repeated impact conditions. Notice that this shift is not equal for all particle sizes. The maximum shift appears to occur at particle sizes around 2000 microns.

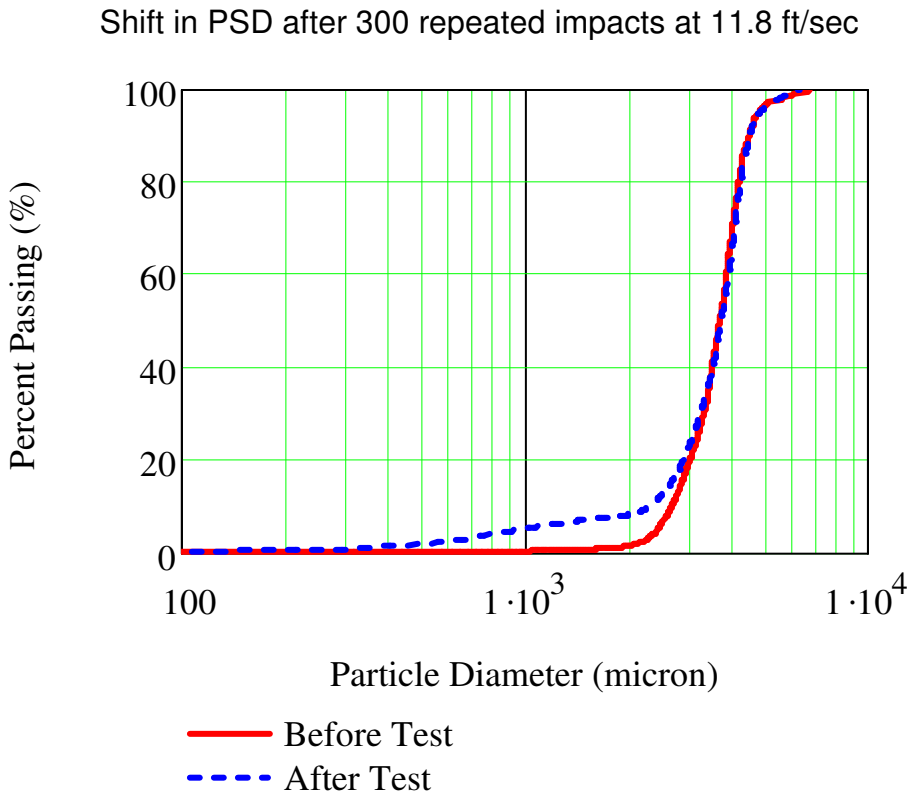


Figure 1. Shift in particle size of bran pearls when subjected to 300 impacts at 11.8 ft/sec impact velocities

Degradation can be better analyzed by looking at the difference between these two curves as a function of particle size (Figure 2). This gives an understanding of which size particles are more prone to breakage. This method of analysis gives slightly more information than a single index; knowledge that is useful in understanding the cause of size degradation in a particular process. Figure 2 describes the expected attrition caused by impact at velocities between 2.9 and 11.9 ft/sec. All of these curves reveal a peak in attrition at a particle size between 1000 microns and 2500 microns, depending on the impact velocity. This peak grows with increasing impact velocity, indicating that impact causes dominant breakage of particles that generate fines that are primarily 2500 microns and smaller. Particle sizes above 2500 microns, therefore, break at a faster rate than they are generated from larger size particles. The lowest impact velocity indicates very little overall attrition, but shows a different behavior than what occurs at higher impact velocities. There appears to be a peak generation of particles around 4000 microns which may be caused by breakage of doublet and triplet bran pearls. This implies a change in the cause of particle size attrition with impact velocity. However, performing the degradation test in this manner still does not provide sufficient information to quantify degradation causes. A more robust method to determine particle breakage mechanisms will be presented later in this paper.

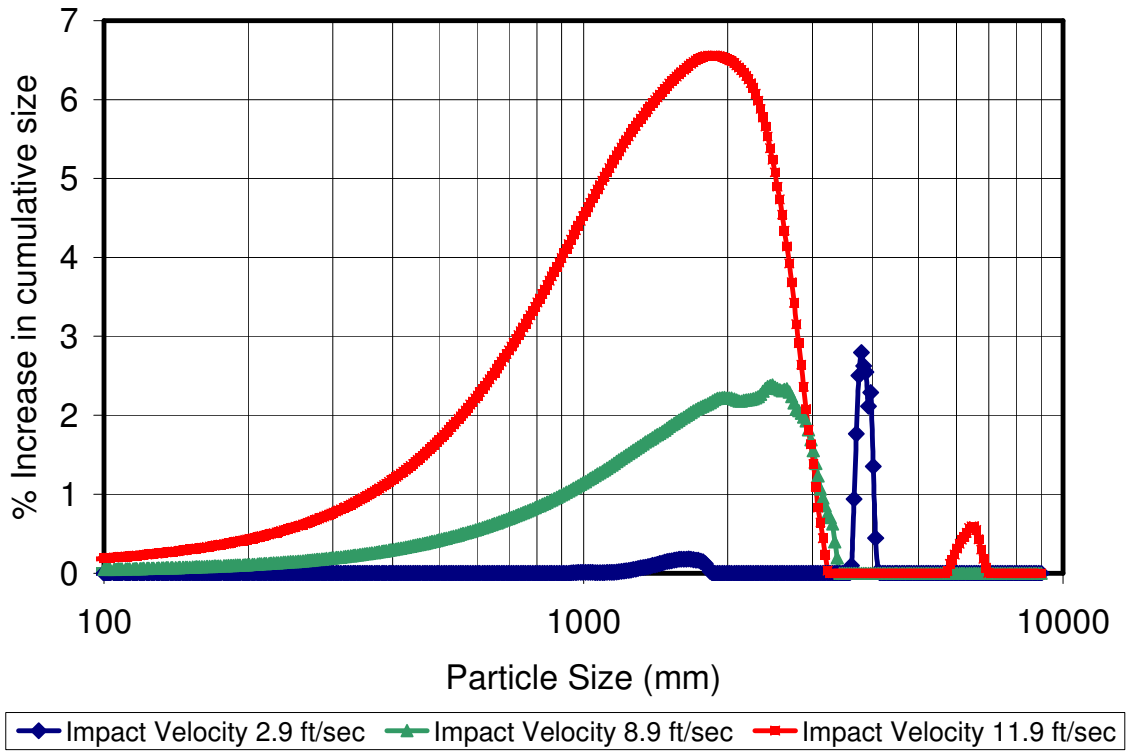


Figure 2. Increase in given cumulative particle size of bran pearls when subjected to 300 impacts at various impact velocities

We have only presented degradation as a function of impact for this material. Thus, this degradation test has applicability to process situations where impact controls particle breakage. It is possible to use this attrition information to estimate the particle size degradation in prescribed process conditions. The first step in this analysis is to determine the critical particle size that represents an index to attrition in the process. For example, suppose that customer acceptance drops off appreciably when the bran pearls have a significant amount of particles less than 900 microns. The amount of increase in particles sizes less than 900 microns could be used as a customer acceptance index to monitor the production process. The information in Figure 2 could then be used to determine particle size degradation as a function of impact velocity for 900 micron particles (Figure 3). However, the process must first be specified and the impact velocities calculated for all parts of the process. Suppose that the process of interest is a rotary drum used for drying. If the drum is 2-foot in diameter, has lifter bars, and operates at 20 RPM with a fill rate of 7%, then the expected maximum impact velocities are around 8.6 ft/sec. However, multiple impacts occur after the initial drop onto the pile from the lifter blades. In addition, not all the material drops the full distance. Taking all this into account and assuming about 100 impact events before material exits the drum, it is estimated that there will be an increase in 900 micron material of about 0.42%. Please note that many processes induce both impact and stress and strain effects on particles. Therefore, a complete degradation process analysis will usually require measurement of size degradation caused by both impact and stress/strain events. Once the process is specified and the stress/strain profiles and impact

conditions in equipment are computed, material degradation can be estimated from the result of degradation tests measured at constant conditions.

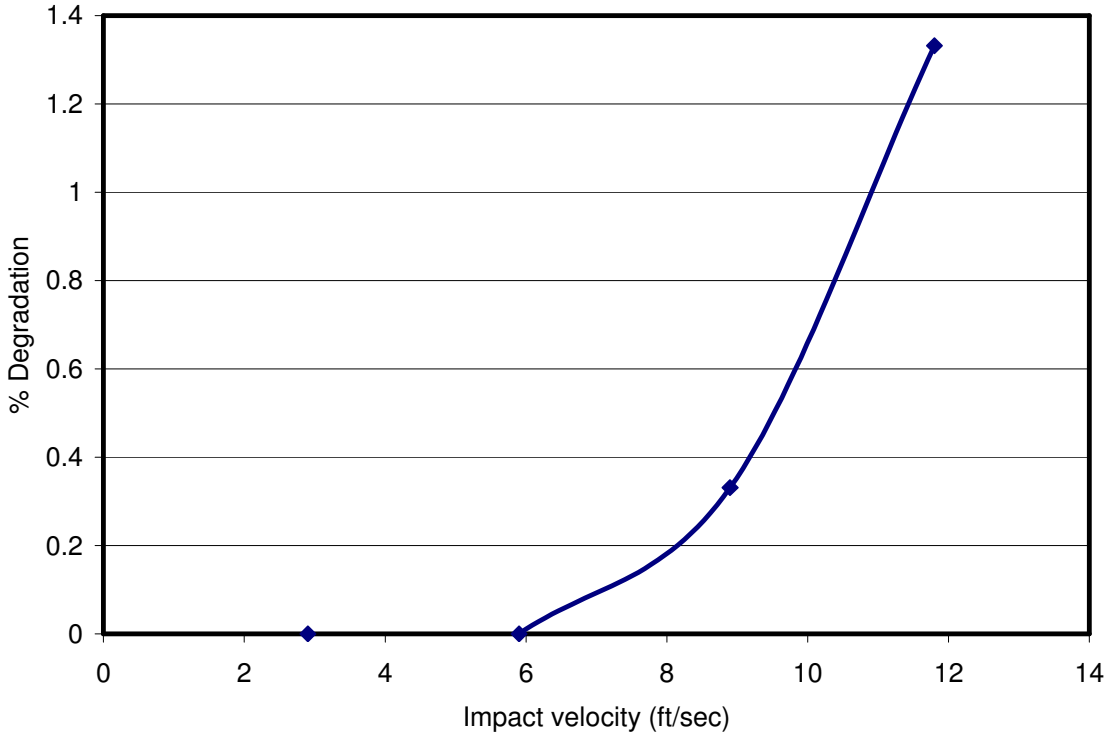


Figure 3. Increase in 900 micron bran pearls when subjected to 100 impacts at various impact velocities

Determining degradation mechanism for bran pearls

Sometimes it is necessary to modify the material to create more robust particles and mitigate degradation. Particles, especially agglomerates, consist of a set of smaller sized particles held together by inter-particle or crystalline forces. The strength of the grain boundaries is one governing factor that limits particle size degradation. The agglomerate structure also plays a primary role in mitigating degradation. To understand this from a theoretical point of view, one requires a model describing the breaking of particles from a structural approach. However, measurement of particle breakage mechanisms is equally important. Therefore, it would be helpful to understand the fundamental mechanisms that create smaller particles from a larger one. As described by Epstein, and Broadbent and Calcott, population balance modeling provides a means of identifying critical particle breakage mechanisms through the computation of breakage selection coefficients [3, 4]. When this is coupled with a structural examination of the agglomerate particle, a powerful tool evolves which allows enhanced particle design. The first step in using this population balance model is to divide the particle size distribution function into regions called particle size bins. These bins are chosen so that an adjacent bin contains particles that are roughly about half the size of the next larger bin. The modeling method monitors the rate of transfer between different particle size bins.

Since the population balance model is at the heart of understanding degradation rates, it is useful to define some parameters and functions that describe breakage, as did Sedlatschek and Bass [7].

- Mass density: $m(x, t)$ & cumulative density: $R(x, t)$
- Breakage distribution function: $B(x, y)$
- Specific breakage rate function: $S(x)$

$B(x, y)$ describes the fraction of mass transported from y to x upon breakage of y.

The breakage distribution function (B) is an indication of where (what particle size bin) the particle fragments end up during breakage. It is often described as a matrix of numbers indicating the fractional distribution of transfer to smaller bin sizes than the original. The numbers in this matrix represent the fraction of material transferred to adjacent particle size bins during processing. The B(x,y) function plays the role of a weighting function or stoichiometric constant in a reaction.

The S(x) function represents the probability that a given particle impact or contact will result in particle breakage. It represents an overall rate of one particle size breaking into any adjacent particles size bins. S functions are the rate constants for degradation from one particle size to all others, as stated by Kapur and Agrawal [5]. Equations governing the rate of transfer between particle size bins can be written for both the cumulative and mass density distributions (see equation 1).

$$\frac{dm_i(t)}{dt} = -S_i m_i(t) + \sum_{j=1}^{i-1} b_{ij} S_j m_j(t), \quad m_i(0) = m_{i0}$$
$$\frac{dR_i(t)}{dt} = -S_i R_i(t) + \sum_{j=1}^{i-1} \left(S_{j+1} B_{ij+1} - S_j B_{ij}\right) R_j(t), \quad R_i(0) = R_{i0} \qquad (1)$$

S-values should correspond to the overall degradation measured from the test method. In general, if the particle size degradation measured from repeated impacts or resulting from stress/strain is low, S-values will also be low. However, S-values give us more specific information since they are specific to a given particle size bin. In general, we must solve the equations above for either the cumulative distribution (R) or the mass distribution (m) and find the rate constants from the solution of these equations. For the purposes of this work we will assume the rate constants are constant and not a function of time. This simplifies the analysis somewhat. However, other assumptions are possible. Consider the work of Bilgili, et al [2].

B functions are also found simultaneously, from the solution of these equations. Since mass cannot be destroyed, the sum of the B functions for any particle size bin must equal one. This solution of the above equations is not a trivial task, but it provide a great deal more information concerning the degradation phenomena experienced by the bulk powder material. Choosing the particle size bins such that each adjacent bin size consists of particles one half the size of the original size bin suggests that movement of material to an adjacent bin would be interpreted as splitting the original particles in half. If the B values indicate a large fraction for the next

adjacent bin size, then fracture of primary particles is occurring. However, if the B values indicate that one or more size bins adjacent to the original size have low fractions followed by large fractions distributed in the finer size bins, then abrasion is likely occurring. Abrasion is the creation of fine particles directly from larger sized particles.

The probability S-function is a value that identifies the likelihood that a collision will result in a breakage of particles. This probability is a function of the size of the particles which typically shows an increasing functionality between the particle size and the probability that a given size particle will break. The solution of the cumulative equations given in equation 1 can be calculated numerically [6] but can also be approximated by the following equations if the rate constants are assumed time invariant [5] .

$$R_i(t) = R_i(0)\exp\left(G_i t + H_i\, t^2/2\right) \tag{2}$$

$$G_i = -S_i + \sum_{j=1}^{i-1} \lambda_{ij} \frac{R_j(0)}{R_i(0)}, \quad H_i = \sum_{j=1}^{i-1} \lambda_{ij}\left(G_j - G_i\right)\frac{R_j(0)}{R_i(0)}, \quad \lambda_{ij} = S_{j+1}B_{ij+1} - S_j B_{ij}$$

The particle size distribution of bran pearls when subjected to repeated impacts at velocities of 17.9 ft/sec was measured at various times to generate the time sequence data required for population balance modeling (Figure 4). The equations listed above were solved with this time sequence PSD data to determine the B(x,y) distribution matrix and the S(x) breakage rate function (Figures 5 and 6).

The S-values represent the overall rate of particle size degradation. Figure 5 indicates that coarse particles around 4300 microns break at a rate that is about eleven times the rate of 2000 micron particles. This indicates a weakness in large particles or bran pearls. The S-value data only indicate overall breakage rate, it does not determine the type of breakage that results from impact. The B-value data contain this information. Consider the data presented in Figure 6.

This data suggest that 45% of material initially in bin #1 (6258 micron) ends up in bin #2 (3284 micron). Thus, 45% of the breakage of these coarse particles is due to fracture where the particles split in half. The remainder of these coarse particles is distributed from bin #3 through bin #7, indicating that some limited abrasion is also occurring with this material. This pattern of a dominant fracture-mechanism fracture with some secondary abrasion also occurs for bin #2 (3284 micron) and bin #3 (1650 micron). However, particles smaller than 904 microns show an abrupt change in breakage mechanism. The primary cause of breakage with these mid-size particles is abrasion, in which large particles become very small particles through knocking off edges and corners. Thus, the population balance model alone gives a quantitative measure of the type of breakage that can occur with bran pearls.

This can be coupled with a structural study of the bran pearls to help determine the particle structures that may be leading causes of size degradation. It is important to point out that the bran pearls are primary particles that are cylindrical with a height-to-diameter ratio of about 1.0

to 1.1. These primary particles are about 4000 micron to 4500 microns with a series of doublets and triplets. The first particle size bin goes from about 4300 to 8300 microns and includes these doublets and triplets. An optical analysis based on the aspect ratio of the original particles indicates that 8.4% of the particles are triplets and 25.3% of the particles are doublets. Optical analysis of the coarse fraction was also performed after subjecting this material to repeated impacts at 17.8 ft/sec. This analysis indicated 3.5% triplets and 21.1% doublets. This gives an overall reduction in doublets and triplets of about 9.1%. The selectivity chart (Figure 6) summarizes B-values from the population balance and indicates that 45% of the material in bin size #1 (6258 microns) transfers to bin #2 (3284 microns). The remainder of the material in bin #1 distributes between bins #3 (1650 micron) and #4 (904 micron) with some trace amounts in the finer bin sizes. When the results from the optical analysis are combined with the population balance model, a clearer picture of the breakage phenomena results. The optical analysis suggests that 9.1% of the 45% total material that moves from bin size #1 to bin size #2 is due to the breakage of doublets and triplets. In other words, 20.9% appears to be due to breakage (fracture) of primary particles in half. The selectivity plot also indicates that all of the material in bin size #2 (3284 micron) ends up as particles smaller than or equal to bin size #4 (904 microns). Almost all of the particles in bin size #3 (1650 micron) end up as particles around (904 microns). The population balance model indicates that particles that are initially 904 microns and 475 microns fall apart into particles that are about 130 microns. This suggests that bran pearls break into individual particles of about 4300 microns which, in turn, fracture into particles around 904 microns. Abrasion then causes these particles to crumble into particles around 130 microns.

A close look at the structure of the particle indicates that a bran pearl is comprised of large flakes that have a D_{50} particle size of about 640 microns (Figure 8). The population balance model suggest that these bran pearls (4500 microns) fracture into smaller bran particles that are around 903 microns average particle size. Once this occurs, smaller particles are knocked off the corners of larger particle during repeated impact to yield fine bran powder (130 microns).

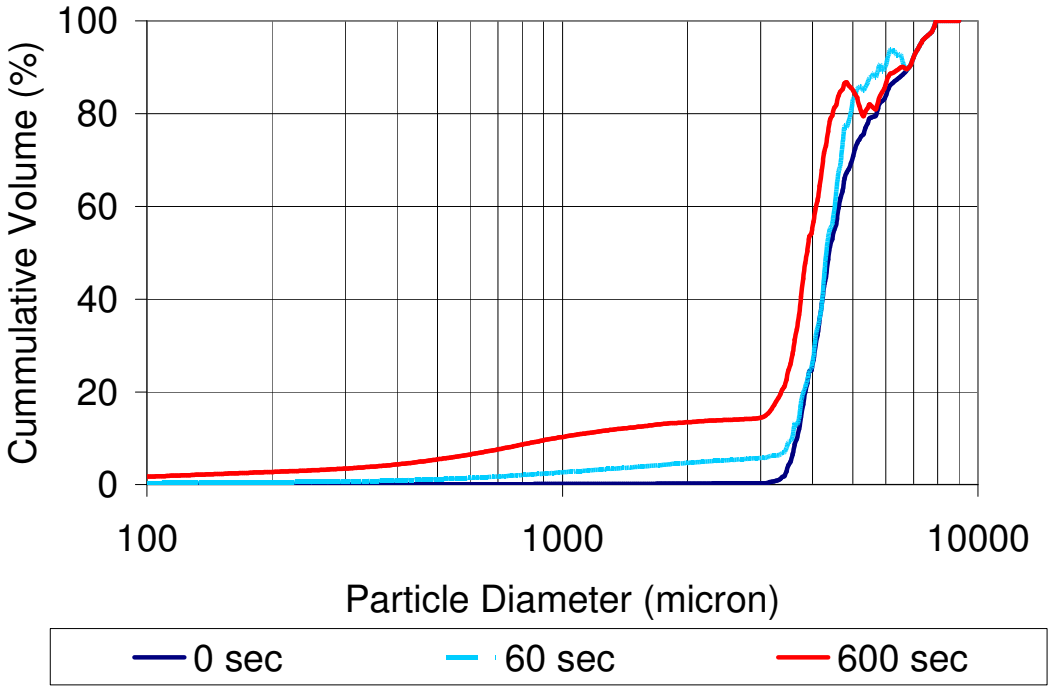


Figure 4. ***Time sequence particle size distribution data for repeated impacts of bran pearls at impact velocities of 17.9 ft/sec***

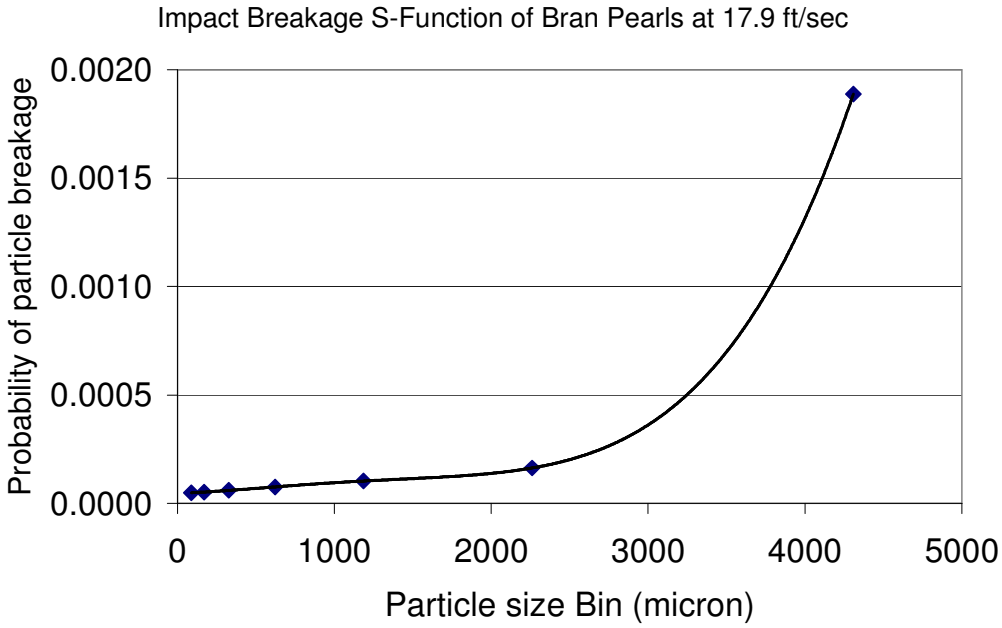


Figure 5. ***S-Values for bran pearls subjected to repeated impacts at 17.9 ft/sec impact velocities***

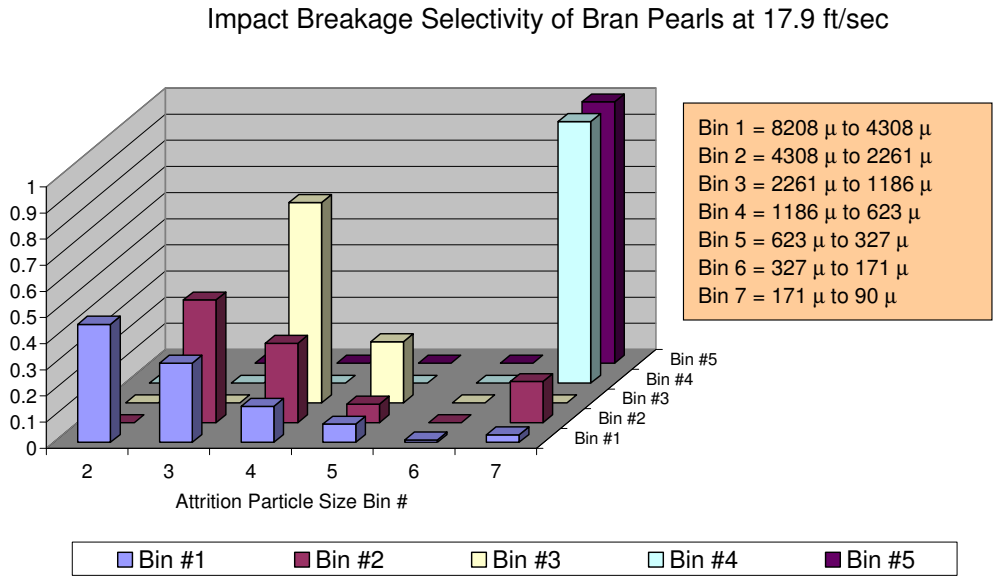


Figure 6. ***B-Values for bran pearls subjected to repeated impacts at 17.9 ft/sec impact velocities***

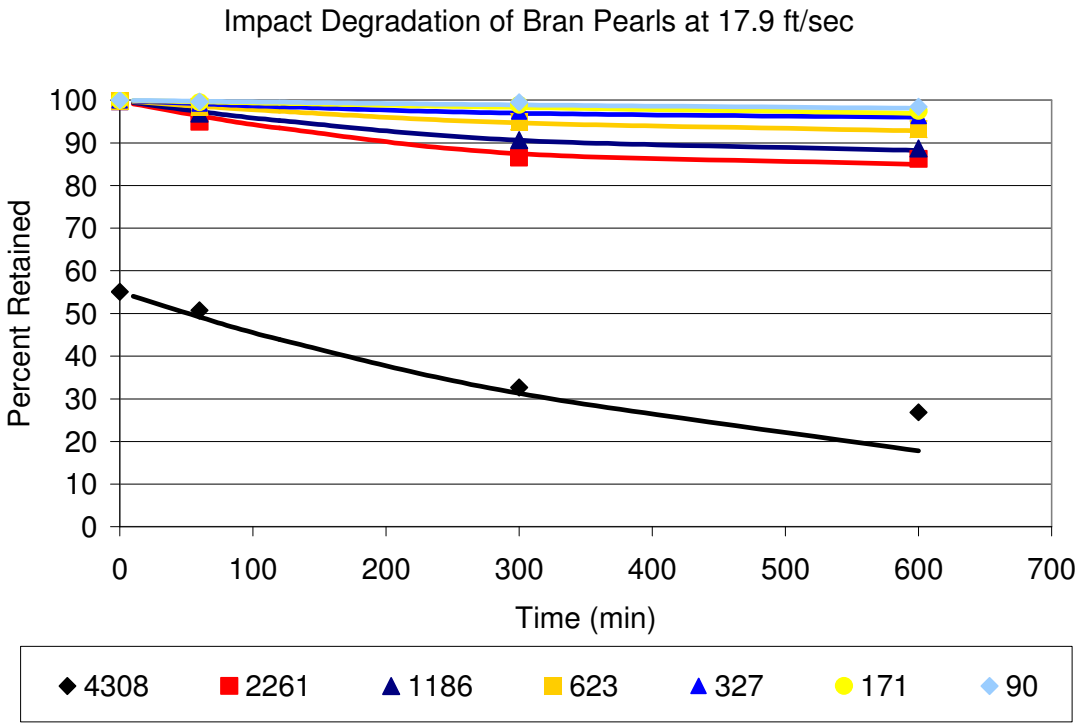
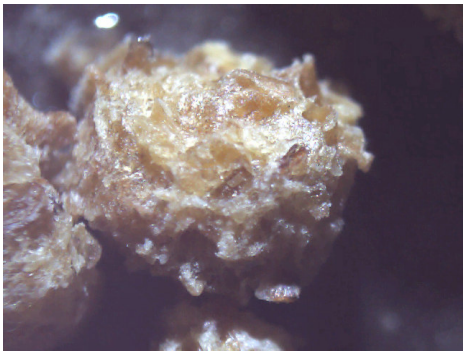


Figure 7. ***Cumulative particle size distributions as a function of time for bran pearls subjected to repeated impacts at 17.9 ft/sec impact velocities***



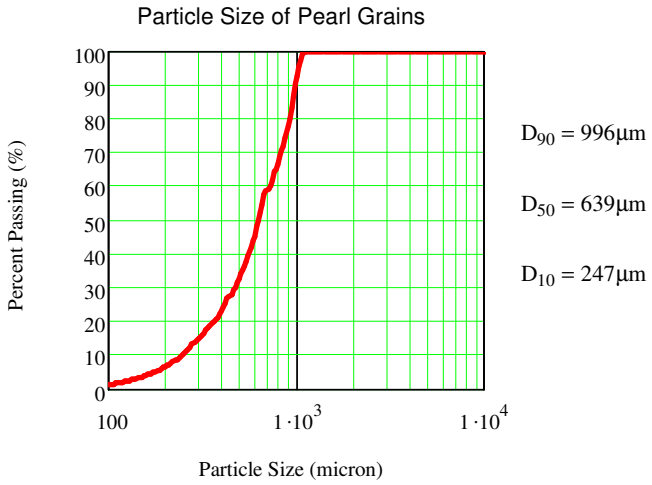


Figure 8. ***Bran pearls showing primary particles about 640***

Determining degradation mechanism for bran fibers

Bran cereal is also formulated in another shape consisting of long extruded particles with a length of 14,000 micron and a diameter of 2100 microns. Particle size degradation was measured after subjecting the material to repeated impacts at 17.8 ft/sec. The data in this case

showed substantial change in particle size within the first 10 seconds followed by a smaller shift in particle size (Figure 9). This trend is also evident in the cumulative particle size distributions plots as a function of impact time (Figure 10). This figure indicates that the amount of particles less than 4300 micron drops 20% within the first 10 seconds of impact. A population balance study of this material suggests that a breakage rate constant independent of time is not sufficient to predict the observed fast size degradation behavior. Thus, the population balance model approach was used to analyze data for the first 60 seconds as well as for data between 60 seconds and 600 seconds. Two different sets of breakage rate constants were obtained for this data.

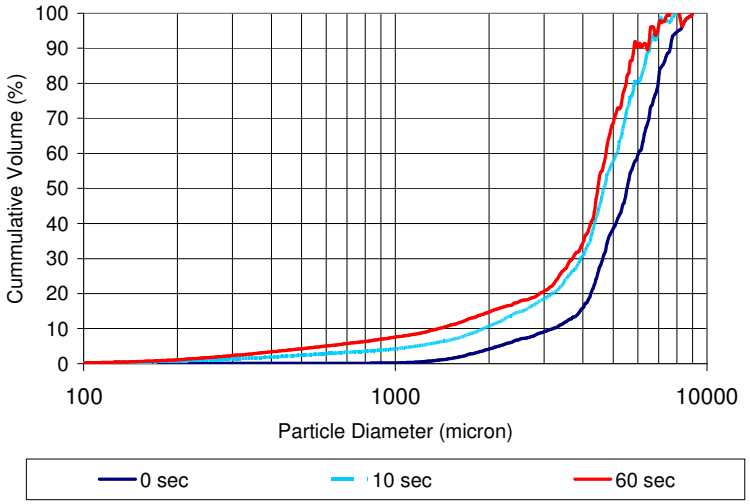


Figure 9. Time sequence particle size distribution data for repeated impacts of bran fibers at impact velocities of 17.9 ft/sec

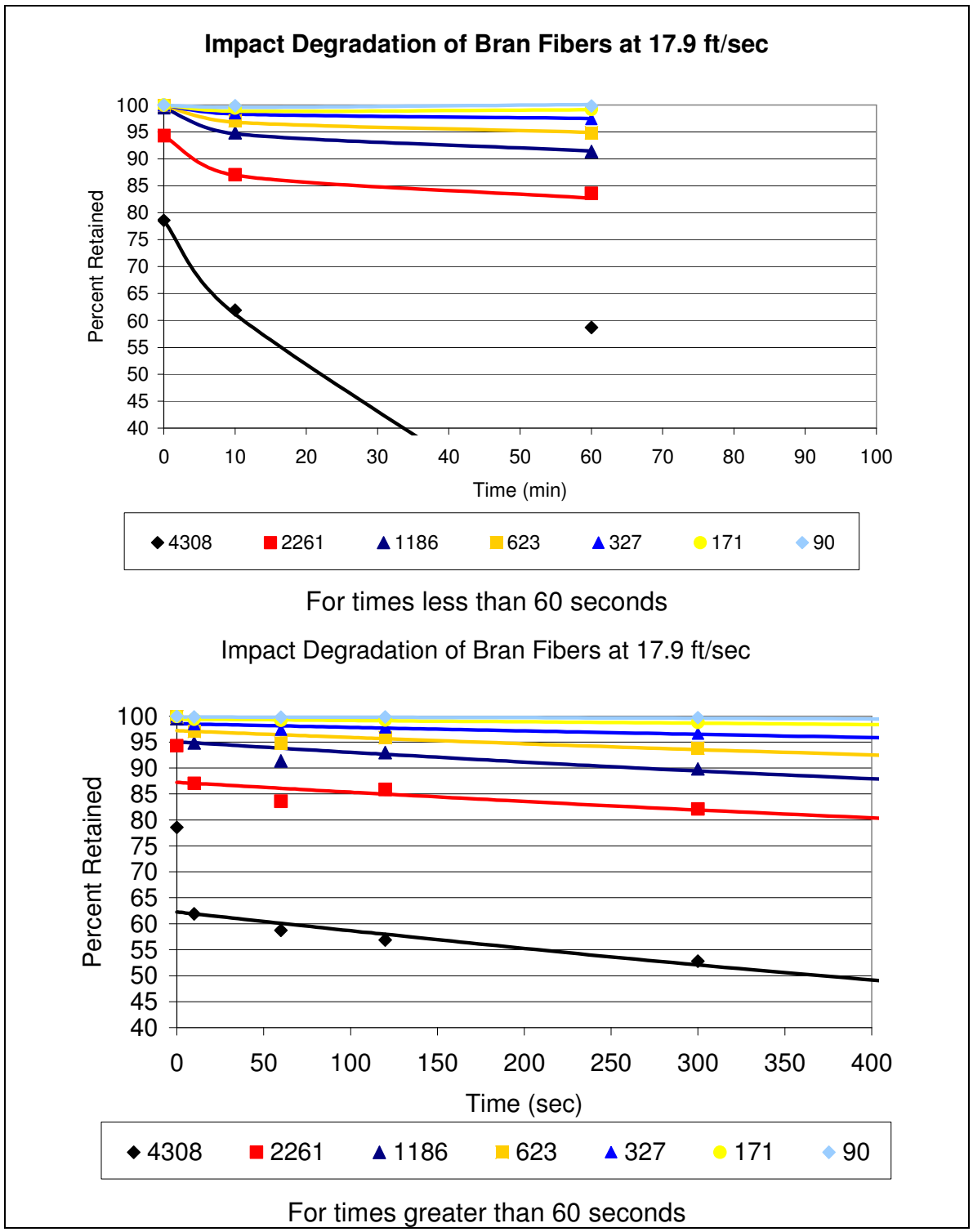


Figure 10. ***Cumulative particle size distributions as a function of time for bran sticks subjected to repeated impacts at 17.9 ft/sec impact velocities***

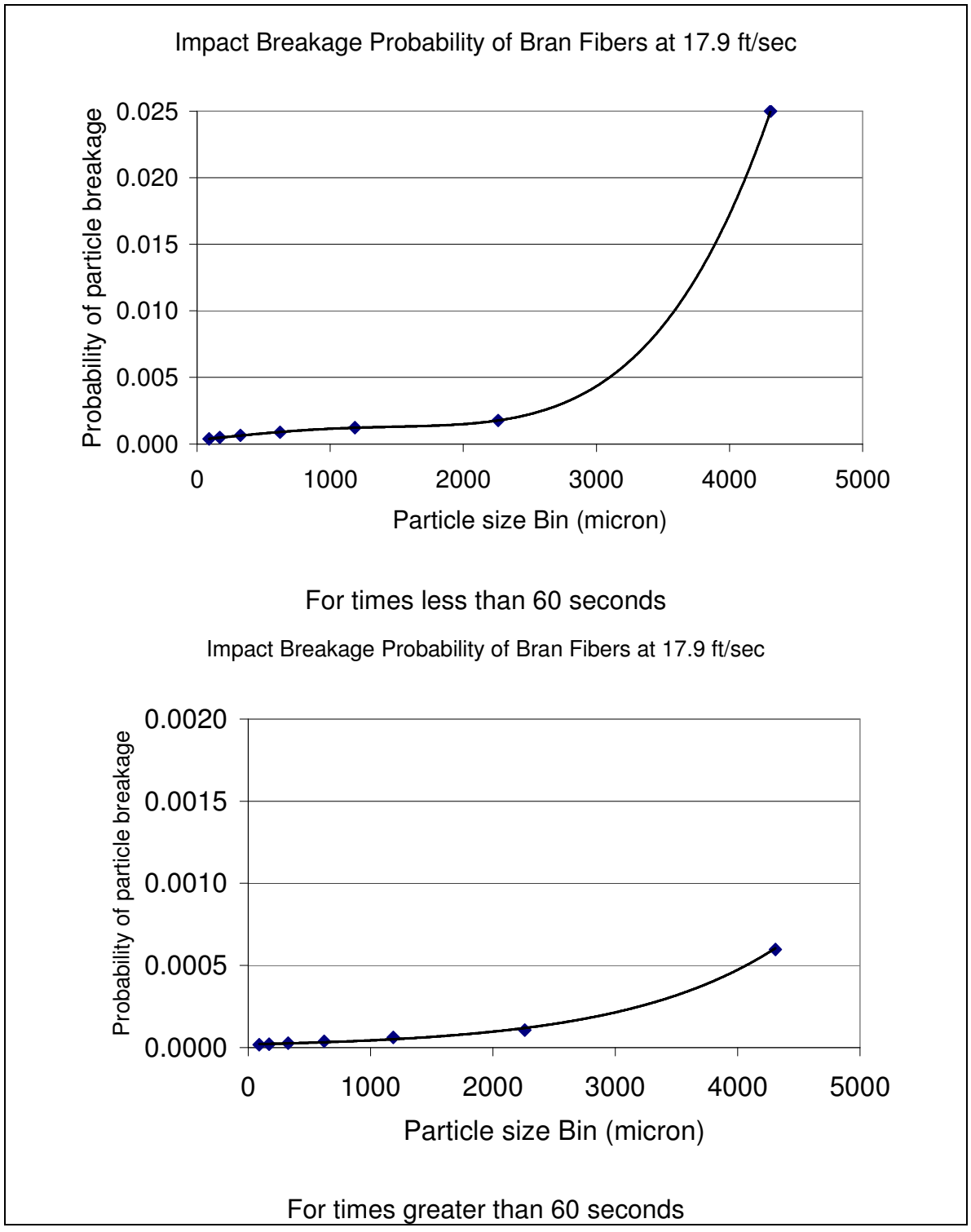


Figure 11. S-Values for bran fibers subjected to repeated impacts at 17.9 ft/sec impact velocities

The breakage rate of large particles occurs very quickly and then slows to a rate that is about 43 times smaller than the initial breakage rate. This same trend persists for smaller size particles where the initial breakage rate is about 20 times the breakage rate observed after 10 seconds of repeated impacts. To aid this analysis, the ratio of the breakage rate S-value curves for the initial breakage and the S-value breakage rate for the long term events were plotted (Figure 12). There is always a reduction in the breakage rate as particles become smaller. This is due to the increase in energy required to produce additional surface area of smaller size particles. However, if the breakage mechanism is the same in the initial stages as compared to the final stages of attrition testing, then the ratio of these two overall breakage rates should yield a constant value. The fact that it does not indicates a change in breakage mechanism that occurs over time. It appears that the minimum overall production of 2200 micron particles is lowest with this material. The larger particles have twice the potential to break during the initial phase of breakage events. Population balance modeling provides a way of quantifying this effect. The breakage selectivity functions B-values for the bran sticks initial as well as long term breakage events were computed. This data is presented in Figure 13 and shows a strong tendency to form particles in the next adjacent bin size during this initial impact phase, indicating fracture of large particles. Thus, the bran sticks tend to break initially by fracture. There is a shift towards abrasion near the end of the degradation testing indicating that after initial fracture particles require repeated impacts to knock off edges. A close look at the bran fiber particles with a large aspect ratio suggests that these particles likely break into two or three pieces due to bending during an impact event to produce particles with an aspect ration closer to one. These more rounded particles then tend to behave similarly to the bran pearls and break because of both fracture and abrasion. There also appears to be a distinct change in breakage mechanism as the particles become finer. This indicates that fracture of large particles occurs followed by abrasion to produce fine bran powder.

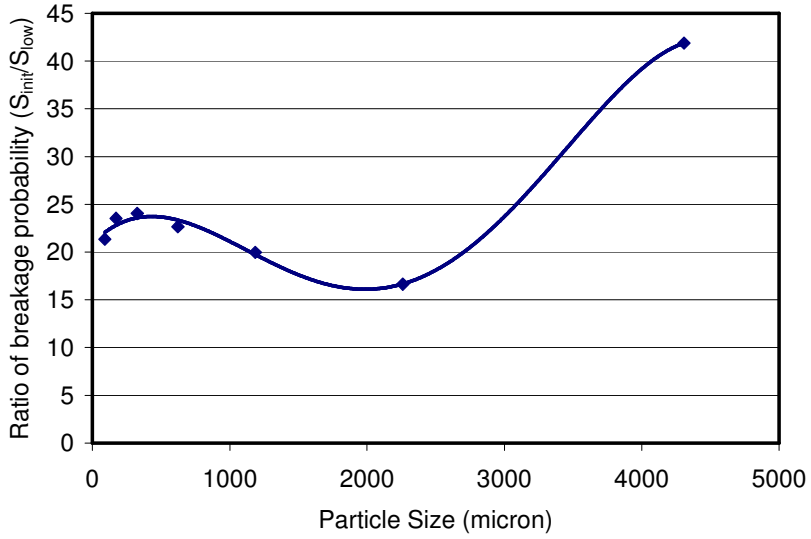


Figure 12. B-Values for bran fibers subjected to repeated impacts at 17.9 ft/sec impact velocities

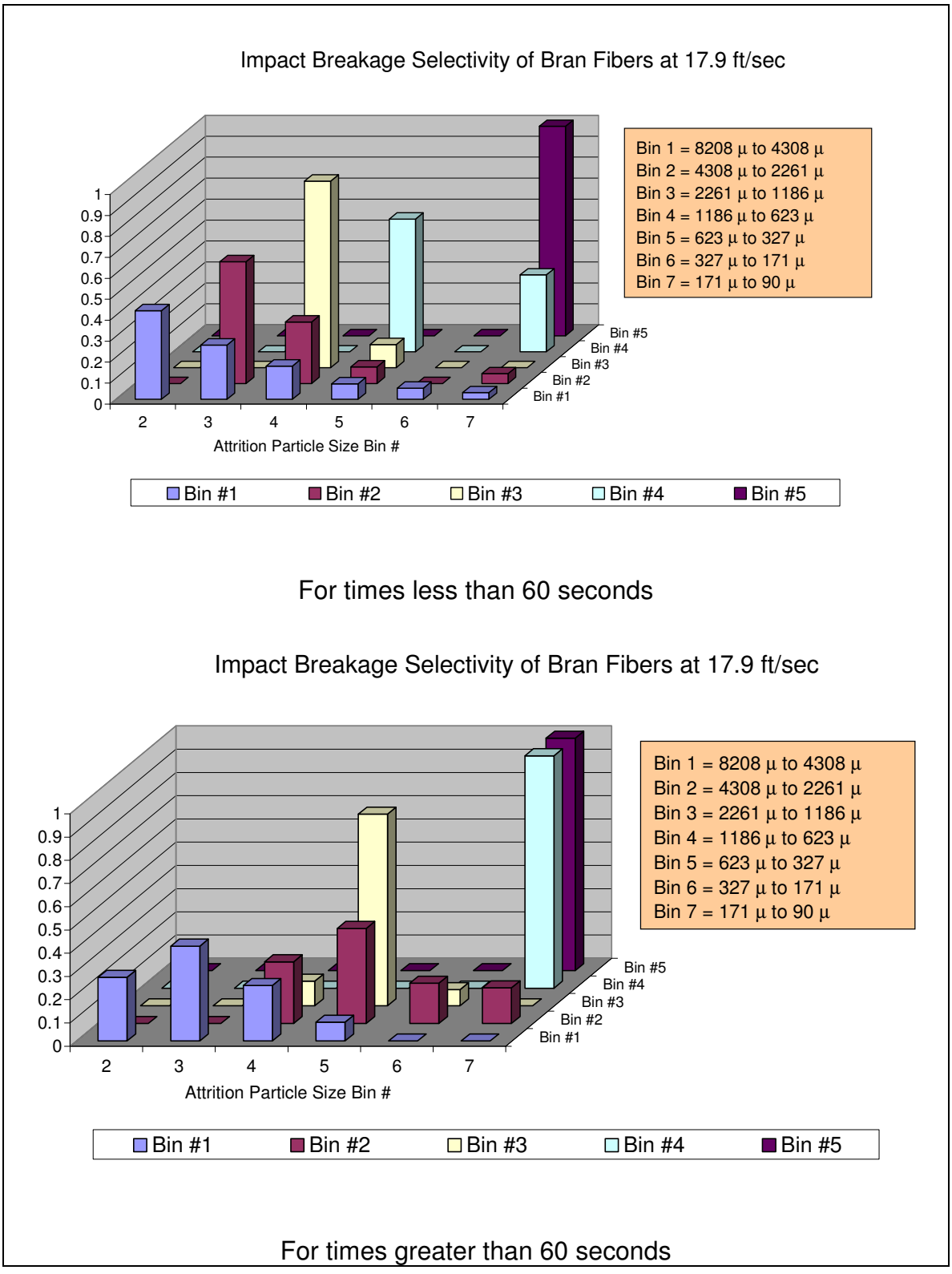


Figure 13. B-Values for bran fibers subjected to repeated impacts at 17.9 ft/sec impact velocities

Nomenclature

B	breakage selectivity constants from the population balance model.
b	differential breakage selectivity constants from the population balance model.
G	internal parameter used to compute the predicted cumulative particle size.
H	internal parameter used to compute the predicted cumulative particle size.
m	differential mass particle size density
R	cumulative particles size.
S	breakage probability function for the particle size bins.
s	differential breakage probability function for the particle size bins.
λ	internal parameter used to compute the predicted cumulative particle size.

Conclusion

If a simple estimate of particle size breakage is required for a typical process, then performing a degradation test with a tester that generates the same type of behavior as in the process can give information necessary to compute an estimate of degradation in the prescribed process. However, simple industrial size degradation tests are often not sufficient to provide information about the cause of particle size degradation of a given material. A more detailed analysis such as the population balance model study is required to help understand the mechanics of particle breakage. When this tool is combined with a structural study of the particle, then the means of preventing breakage becomes evident and provides guidance to create more robust particles. In the case of bran pearls, the main breakage mechanism for large particles was fracture to eventually form particles about 900 microns in diameter. These larger particles then break down by attrition to form fine bran powder (130 microns). If the process that produces these particles could be changed to create a stronger bond between the individual 900 micron bran particles, then these agglomerates would be more robust. Conversely, the bran fibers which consists of large aspect ratio material initially fractures rapidly into particles that are half or one third the size of the original particles. These smaller particles then undergo both fracture and abrasion to generate fines. The maximum rate step is the production of mid-size particles due to the initial fracture of these long particles. In this case, the particle should be designed with some flexibility to withstand bending moments that cause initial fracture of large bran fiber particles. Population balance modeling then provides a powerful means of guiding product design.

References

1. Barletta, Blas J. and Barbosa-Cánovas, Gustavo V., "An attrition index to assess fines formation and particle size reduction in tapped agglomerated food powders," *Powder Technology*, Volume 77, Issue 1, October 1993, pp 89-93

2. Bilgili E. and Scarlett B., "Nonlinear effects in particulate processes," *Nonlinear Analysis*, Volume 63, Issues 5-7, 30 November 2005 to 15 December 2005, pp e1131-e1141

3. Broadbent and Calcott, J. *Inst. Fuel,* Vol 30, 1957, pp 13-15

4. Epstein, B., “Logarithmico – Normal Distribution in Breakage of Solids”, *Ind. Eng. Chem.* Volume 40, pp 2281-1191

5. Kapur, P.C. and Agrawal P.K., “Approximate solutions to the discretized batch grinding equation,” *Chemical Engineering Science*, Volume 25, Issue 6, 1970, pp 1111-1113

6. Reid, K.J., “A solution to the batch grinding equation,” *Chemical Engineering Science,* Volume 20, 1965, pp 953-963

7. Sedlatschek K., Bass L., “Contribution to the theory of milling processes,” *Powder Metal,* Bulletin 6, 1953, pp 148-153