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MRS/SAMPE ABSTRACT

CERAMIC GRANULE STRENGTH VARIABILITY AND COMPACTION BEHAVIOR

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Diametral compression strength distributions and the compaction behavior and of irregular shape 150-200 μm ceramic granules and uniform-size 210 μm glass spheres were measured to determine how granule strength variability relates to compaction behavior of granular assemblies. High variability in strength, represented by low Weibull modulus values ($m<3$) was observed for ceramic granules having a distribution of sizes and shapes, and for uniform-size glass spheres. Compaction pressure data were also analyzed using a Weibull distribution function, and the results were very similar to those obtained from the diametral compression strength tests for the same material. This similarity suggests that it may be possible to model granule compaction using a weakest link theory, whereby an assemblage of granules is viewed as the links of a chain, and failure of the weakest granule (i.e., the weakest link) leads to rearrangement and compaction. Additionally, with the use of Weibull statistics, it appears to be possible to infer the variability in strength of individual granules from a simple pressure compaction test, circumventing the tedious task of testing individual granules.

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DISCLAIMER

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ABSTRACT

Diametral compression strength distributions and the compaction behavior and of irregular shape 150-200 μm ceramic granules and uniform-size 210 μm glass spheres were measured to determine how granule strength variability relates to compaction behavior of granular assemblies. High variability in strength, represented by low Weibull modulus values ($m<3$) was observed for ceramic granules having a distribution of sizes and shapes, and for uniform-size glass spheres. Compaction pressure data were also analyzed using a Weibull distribution function, and the results were very similar to those obtained from the diametral compression strength tests for the same material. This similarity suggests that it may be possible to model granule compaction using a weakest link theory, whereby an assemblage of granules is viewed as the links of a chain, and failure of the weakest granule (i.e., the weakest link) leads to rearrangement and compaction. Additionally, with the use of Weibull statistics, it appears to be possible to infer the variability in strength of individual granules from a simple pressure compaction test, circumventing the tedious task of testing individual granules.

KEY WORDS: Compaction, Granule, Weibull distribution

1. INTRODUCTION

Compaction of powders and granular materials has considerable commercial importance in the materials, pharmaceuticals, food, and mineral processing industries. For example, in the powder metallurgy and ceramic manufacturing industries, powder compaction techniques are commonly used to form components. Compaction of granulated materials is particularly important in ceramic component manufacturing, as submicron ceramic powders are often granulated (e.g., by spray drying) with an organic binder to improve handleability and flow. Increasingly better understanding of the compaction process is desired to fabricate dry-pressed parts with more uniform density, and improved performance and reliability.

1.1 Compaction Behavior of Granular Assemblies The compaction behavior of powders and granular assemblies is often summarized by plotting the relative density of the compact vs. the log of the compaction pressure as shown in Figure 1. This method of representing compaction data provides a semi-quantitative measure of granule strength, and is also useful as a tool to determine the pressure necessary to achieve a given density. When plotted as relative density vs. log compaction pressure, compaction data tend to exhibit linear regimes that are attributed to different compaction mechanisms. Densification in the first linear regime is attributed to rearrangement without fracture. Experimentally, it is often observed that this regime has a slope of ~0.003-0.005 (where slope = $[\rho_2-\rho_1]/[\log P_2-\log P_1]$). The low slope indicates that very little compaction occurs, and that only a small percentage of the void space is removed during rearrangement. In the second linear regime, which often exhibits a slope of ~0.2, densification is attributed to deformation and fracture in conjunction with further

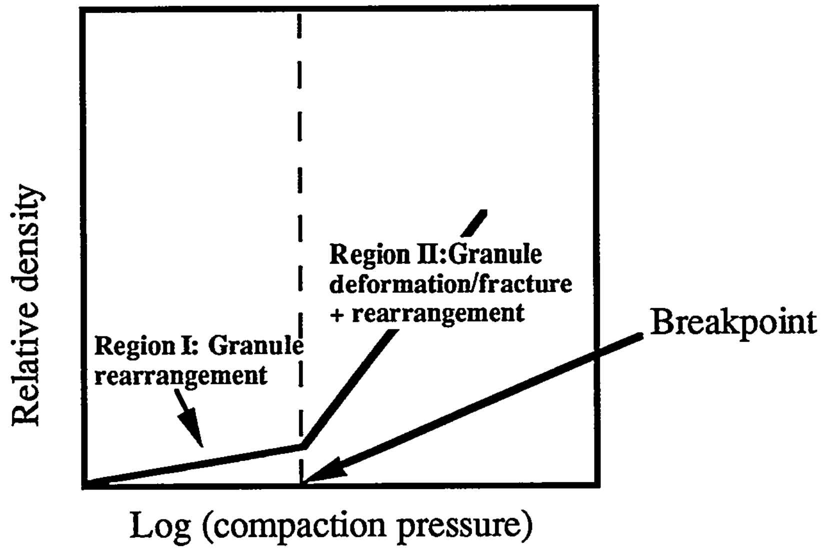

Figure 1. Compaction behavior of ceramic granules plotted in terms of relative density vs. log compaction pressure. Compact densification occurs by granule rearrangement without fracture in Region I. In Region II compact densification occurs by granule deformation and fracture in conjunction with rearrangement. The intersection of the two linear regimes is known as the breakpoint.

rearrangement. There also appears to be another characteristic slope at higher pressures, but compaction behavior at higher pressures has not been as thoroughly characterized, and is notwell understood. Moreover, because of compact ejection problems and excessive die wear, high pressures are usually undesirable in ceramic powder forming; consequently, the high pressure regime is not as relevant and will not be discussed further.

Best fit lines are often drawn through the data in the first two linear regimes of the compaction plot, the intersection of which is known as the breakpoint. The breakpoint defines the pressure at which the compaction mechanism changes from rearrangement to a combination of deformation, fracture, and rearrangement. The breakpoint has been used as a semi-quantitative indicator of granule strength or yield point. Although the breakpoint has been generally thought of as the average strength of the granules, recent work demonstrates that it is more representative of the lower end of the range of granule strengths.(1)

1.2 Characterizing Compaction in Granular Assemblies By characterizing and understanding the compaction behavior of granular materials, it may be possible to better understand how to fabricate uniform green density ceramic bodies (e.g., by dry pressing) that sinter to high uniform density. In an ideal case, this would be accomplished by analyzing a simple compaction diagram constructed with compaction data from a granular assembly; however, while the breakpoint and the Region II slope in a compaction diagram provide some general information on compaction, a more detailed analysis is necessary to better understand and control the compaction process.

Before further consideration of the compaction diagram and how it relates to the rearrangement and subsequent deformation and fracture behavior of the granules, it is important to recognize that other factors can affect the compaction response. One such factor is the stress distribution in the die. If the aspect ratio of the compact is large, there will is large stress gradient from the top to the bottom of the die; consequently, the granules at the bottom of the die are subjected to much lower stresses than those at the top. As a result of the stress gradient, which is due to die wall friction (2), higher applied pressures must be exerted to deform and fracture the granules at the bottom of the die. This gradient changes the slope of the second linear regime to a lower value, as higher forming pressures must be exerted to achieve the same overall degree of densification throughout the compact. The effect is illustrated in Figure 2, which shows compaction behavior for compacts of TiO_2 granules with three different aspect ratios. Both

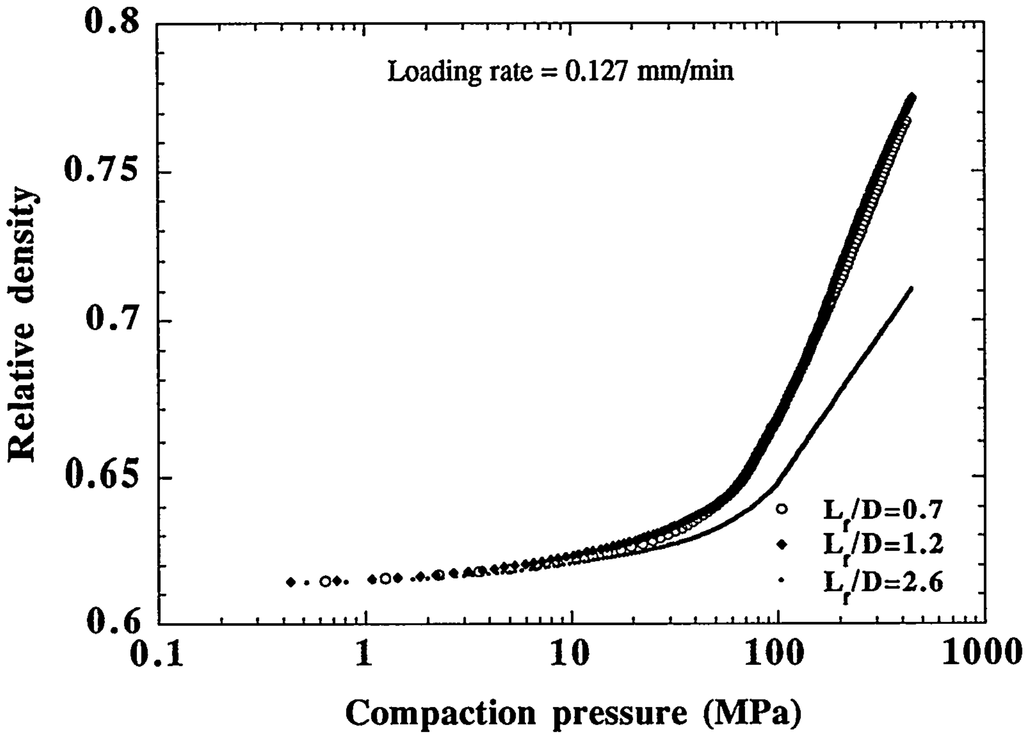

Figure 2. The effect of compact aspect ratio (L_f/D) on the slope of Region II compaction behavior for rutile (TiO_2) granules.

lower slope and lower endpoint density are observed for $L_f/D > 2.6$ (where L_f = compact height after pressing, and D = compact diameter). The effects of aspect ratio on granule compaction can be eliminated by pressing a low aspect ratio compact of $L_f/D \leq 1$. (3)

The compaction behavior of a granular assembly can also be affected by compaction rate. (3) As observed with increasing compact aspect ratio, increasing the compaction rate changes the slope of the second linear regime to a lower value. The effect can be minimized by using slow compaction rates.

Non-uniform die filling can also affect compaction behavior. To simulate and evaluate the effects of non-uniform die filling, Onoda(4) performed compaction experiments using a cylindrical die that was deliberately filled with granules in a convex arrangement as shown in Figure 3. Onoda tested a series of soft, medium, and hard Al_2O_3 granules, where granule hardness (i.e., deformability) was varied by modifying the organic binder system. The three different systems were compacted at a common pressure, and then green density was measured as a function of distance across the compact. Hard granules produced a uniform, but low green density compact. Medium granules produced a uniform, moderate density green compact. Soft granules produced a high average green density, but with an appreciable gradient in density across the compact. Onoda's results indicate that the effects of non-uniform die filling on granular assembly compaction can be minimized or eliminated by using granules that are hard (i.e., strong) enough to survive low pressure rearrangement in Region I of the compaction diagram before higher pressure deformation and fracture.

Therefore, by conducting tests on hard granules using slow compaction rates and small aspect ratio compacts, the effects of non-uniform die filling, compaction rate, and compact aspect ratio on the compaction behavior of a granular assembly can be minimized. (3) By following these guidelines, it should be possible to design and conduct compaction tests with uniform

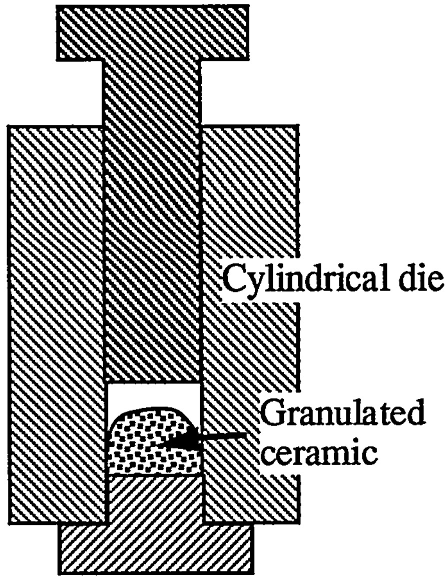

Figure 3. A cylindrical die was deliberately filled with granules in a convex arrangement.

macrostress transmission conditions to determine the effects of the variability in the strength of individual granules directly from simple compaction diagrams.

1.3 Relating Granule Fracture to Granular Assembly Compaction One key to better understanding and controlling compaction in granular materials may lie in the individual granules that make up the compact. Granular materials compaction is undoubtedly intimately related to the deformation and fracture of the individual granules in the compact; therefore, characterizing the fracture strength of individual granules and relating it to the compaction behavior of granular assemblies should provide a better understanding compaction. The objective of this study was to characterize the strengths of individual ceramic granules and the compaction behavior of granular assemblies of the same ceramic materials to determine how granule strength relates to granule fracture and compaction in granular assemblies. A Weibull distribution function(5) based on the concept of weakest link failure, which has been found to be valid for describing ceramic strength data, will be used to analyze and relate the two

2. EXPERIMENTAL PROCEDURE

2.1 Materials Diametral compression strength tests, and granular assembly compaction tests were conducted on lead magnesium niobate-lead titanate (PMN-PT) granules, and on uniform-diameter 210 µm glass spheres. The PMN-PT granules were 150-200 µm in size and were irregularly shaped. Unlike typical spray-dried ceramic granules, all of the PMN-PT granules in this study were heated to sufficiently high temperatures to remove all organic materials. The PMN-PT granules were heat-treated at temperatures of 800, 900, 1000, 1100 and 1200°C, and all had undergone some degree of densification.

2.2 Diametral Compression Strength Tests Granule strengths were measured using diametral compression (DC) tests in which individual granules of diameter d, were mechanically loaded at 0.127 mm/min between platens on a mechanical testing loadframe until fracture at a load of P_{max}. Granule tensile strength, σ_t, was calculated using the equation of Hiramatsu and Oka (6) commonly used in numerous studies.(7,8,9)

$$\sigma_t = \frac{2.9\ P_{max}}{\pi d^2} \qquad [1]$$

2.3 Compaction Tests Compaction experiments were conducted in a 3.2 mm diameter cylindrical steel die at ambient temperature and humidity, with no die lubrication. Three or more tests were conducted on each sample. Tests were conducted on granular assemblies compacted to final aspect ratios, L_f/D, less than 1.0 at 0.127 mm/min. Relative density as a function of compaction pressure was determined using the granule mass, initial die fill height, and die plunger displacement. Corrections for loadframe and fixture compliance were made following the details found in Reference 1. For analysis, relative density and compaction pressure were plotted as relative density vs. log of the compaction pressure.

3. RESULTS AND DISCUSSION

3.1 Granule Strength Weibull plots produced from the measured strengths and strength distributions of the PMN-PT granules are shown in Figure 4. A Weibull parameter, m, of ~3 was determined from the slope of the data in the Weibull plot. Similar results have been obtained for rutile (TiO_2) granules. The Weibull parameter describes the variability of the granule strength distribution, with variability increasing with decreasing m. The granular materials in this study exhibit large variability in strength, and even greater strength variability than typical ceramics and glasses (m = 5-15).

The very low m-values of the granular materials examined requires further discussion. Strength variability, as described by Weibull statistics, is assumed to be related to a variation in the flaw size. Additionally, Weibull parameters are usually calculated for bodies that are the same size and shape. As shown in Figure 5, the granules examined here have irregular shapes, and range in size from 150 and 200 µm,; consequently size and shape variability may account for part of the large variability observed in granule strength. Also, with irregular shape granules under point loading, stress concentrations due to edges and points may increase the variability in the measured strength.

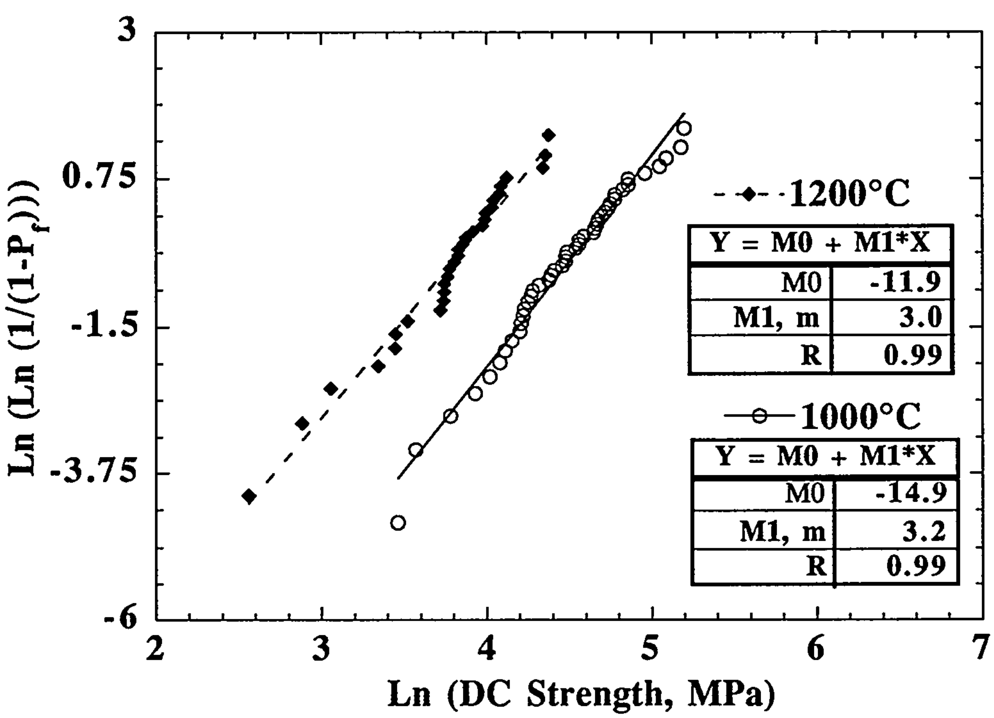

Figure 4. Weibull plots for PMN-PT granules heat-treated at 1000 and 1200°C. For clarity Weibull data for PMN-PT granules heat-treated at 800, 900, and 1100°C are not included in the plot, but low Weibull moduli were also determined for these materials.

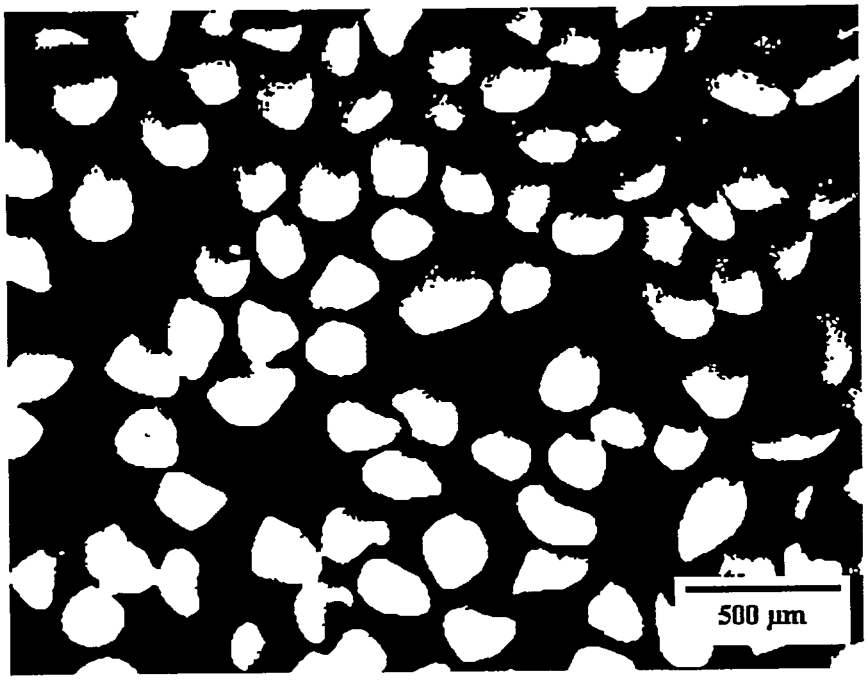

Figure 5. Optical micrograph of PMN-PT granules heat-treated at 1000°C. Their irregular size and shape may contribute to the high variability in granule strength.

To eliminate the effects of shape and size factors, the strength distribution for uniform-size 210 µm diameter glass spheres (Figure 6) was measured. The Weibull results for the glass spheres are shown in Figure 7. Surprisingly, glass spheres with uniform shape and size also exhibited very large strength variability. This result suggests that, even if uniform loading is achievable (if all granules have a similar number of nearest neighbors and applied stress), not all of the granules will break at the same stress. The reason for the high variability in strength of the glass spheres has not been determined, but one possibility is that a large range in flaw sizes may be present.

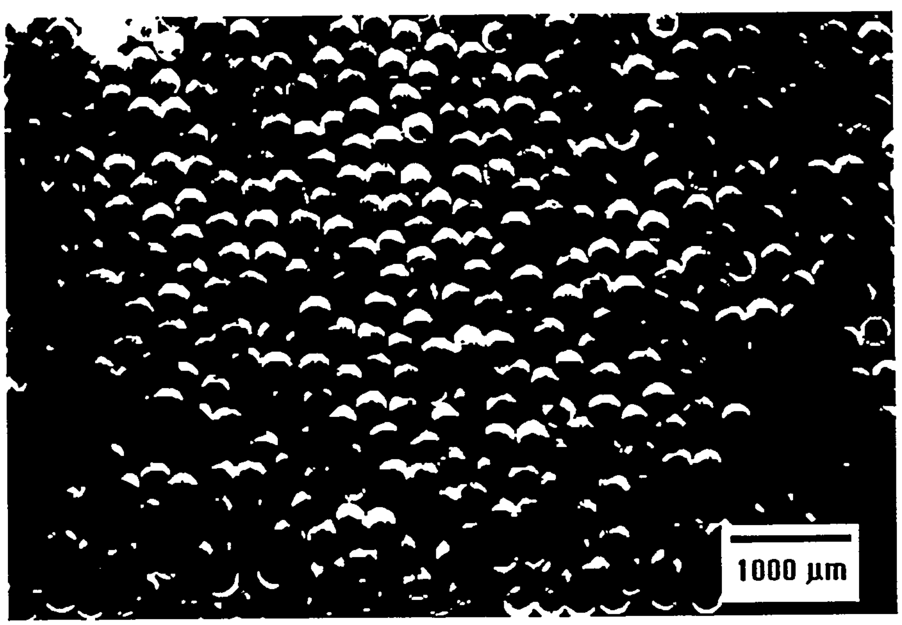

Figure 6. Scanning electron micrograph of uniform-size 210 µm diameter glass spheres.

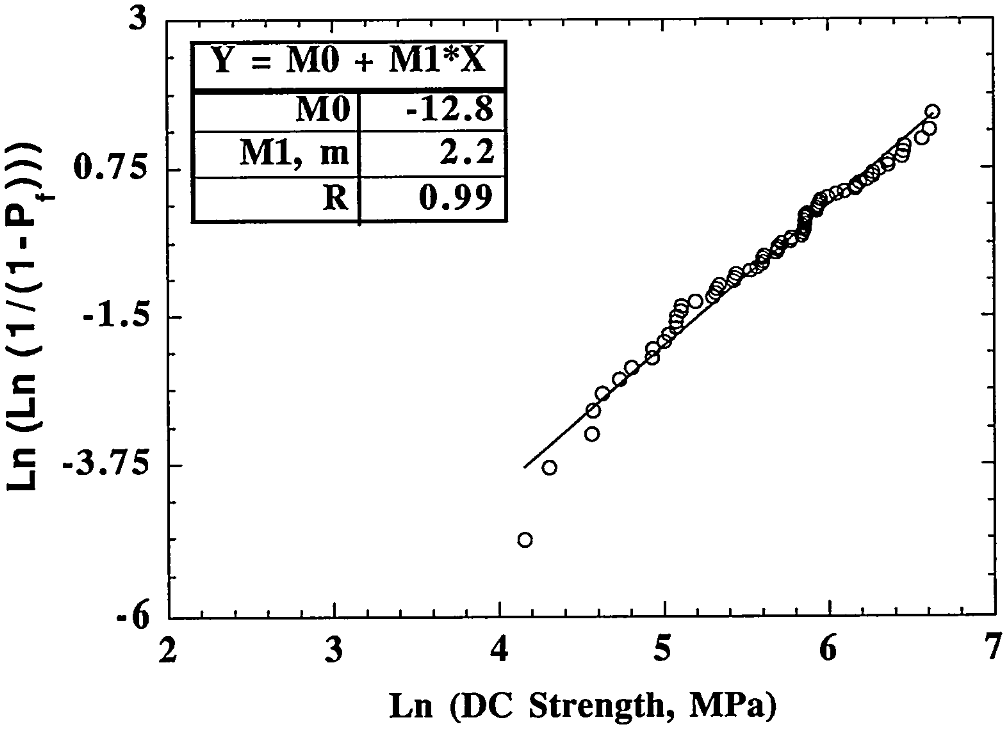

Figure 7. Weibull results for the uniform-size 210 μm glass spheres.

The high variability in strength determined for the PMN-PT granules could indicate a short-coming of Weibull statistics in analyzing granule fracture. It may be possible that Weibull statistics are not directly applicable to test specimens in which the flaw size is large relative to the specimen size. Weibull parameters for ceramics are typically determined from bend tests on specimens that are 3 mm x 4 mm x 50 mm, with typical flaw size ranges from 10 to 100 μm. In such cases, the flaw size relative to the width of the tensile surface is 0.0333 to 0.0033. In the diametral compression tests conducted on individual granules, the flaw can be on the order of the grain size, and a much larger ratio of ~0.1 is possible. Consequently, some refinements may be required to analyze granule fracture using Weibull statistics.

3.2 Granular Assembly Compaction Compaction plots for the PMN-PT granules heat-treated at 800, 1000, and 1200°C are shown in Figure 8. The Region II slopes of the different materials are shown on the plot. All of the materials had similar slopes in Region II of the compaction plot. The similarity in slopes may be related to the fact that all of the individual granules of the materials tested have similar variability in strength, as evidenced by their similar m values.

3.3 Relating Granule Fracture to Granular Assembly Compaction Relating granule fracture strength to the compaction behavior of a granular assembly requires a closer look at the compaction data, and a means of correlating compaction response to strength variability. During compaction, granules in an assembly can be considered as links in a chain. When the weakest link (granule) breaks, the rest of the granules are able to rearrange, and the overall density of the compact increases. This model is similar to the weakest link concept of Weibull statistics. As such, with increasing pressure after Region I, data in a pressure compaction plot represent the fracture of progressively stronger granules in the assemblage of granules. In the linear regime in Region I, no fracture is occurs, and in Region II, many of the granules are shielded from point loading by previously deformed and fractured granules; therefore, to focus on the individual granule fracture events during compaction of the granular

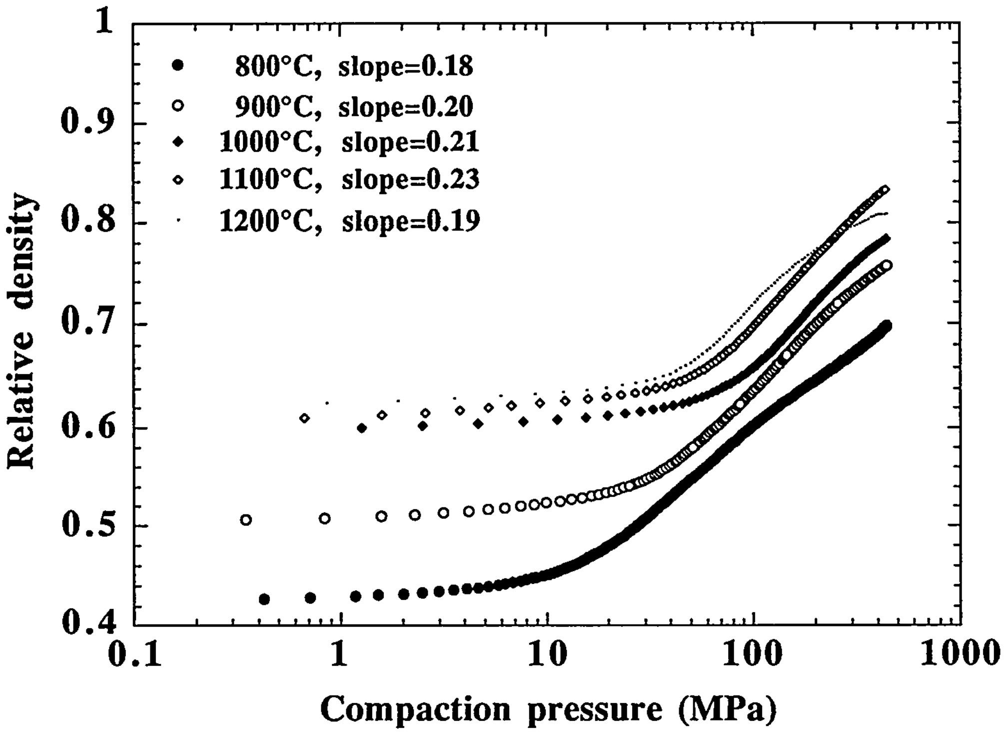

Figure 8. Compaction plots for PMN-PT granules fired at different temperatures. The slopes noted on the legend refer to the second linear (Region II) of the compaction curve.

assembly, only the data in the non-linear region between the two linear regimes in Regions I and II should be used. With these guidelines in mind, compaction data were examined to test the idea that pressure data, at least over the range of stresses where granules break under point loading, represent the fracture of individual granules. The compaction data for the 1000°C heat-treated PMN-PT granules are presented in a Weibull plot in Figure 9. The plot appears similar to the Weibull plot for the individual granule strength data from diametral compression tests (Figure 4), with similar values of m and σ_o. Thus, it appears that the strength variability of individual granules in a compact can be determined directly from the data used to produce the compaction plot. That is, the compaction test appears to provide a simple means to measure the variability in strength of a granular material, circumventing the tedious task of testing individual granules.

4. CONCLUSIONS

Diametral compression tests showed that both irregular shape and size PMN-PT granules and uniform shape and size glass spheres exhibit similarly low Weibull moduli (m<3), which represent very broad strength distributions. The low m-values may reflect the large relative flaw sizes in the materials tested, and suggests that some refinements may be needed to apply Weibull statistics to analyze granule fracture. Compaction pressure data in the non-linear regime between the two linear regimes in Regions I and II of the compaction diagram were also analyzed as Weibull data. The compaction pressure Weibull plots and the Weibull parameters determined from these plots were very similar to those obtained from diametral compression strength tests conducted on individual granules of the same materials. The similarities indicate that the strength variability of individual granules in a compact can be determined directly from the data used to produce the compaction plot. That is, the compaction test may provide a simple means to measure the variability in strength of a granular material, circumventing the tedious task of

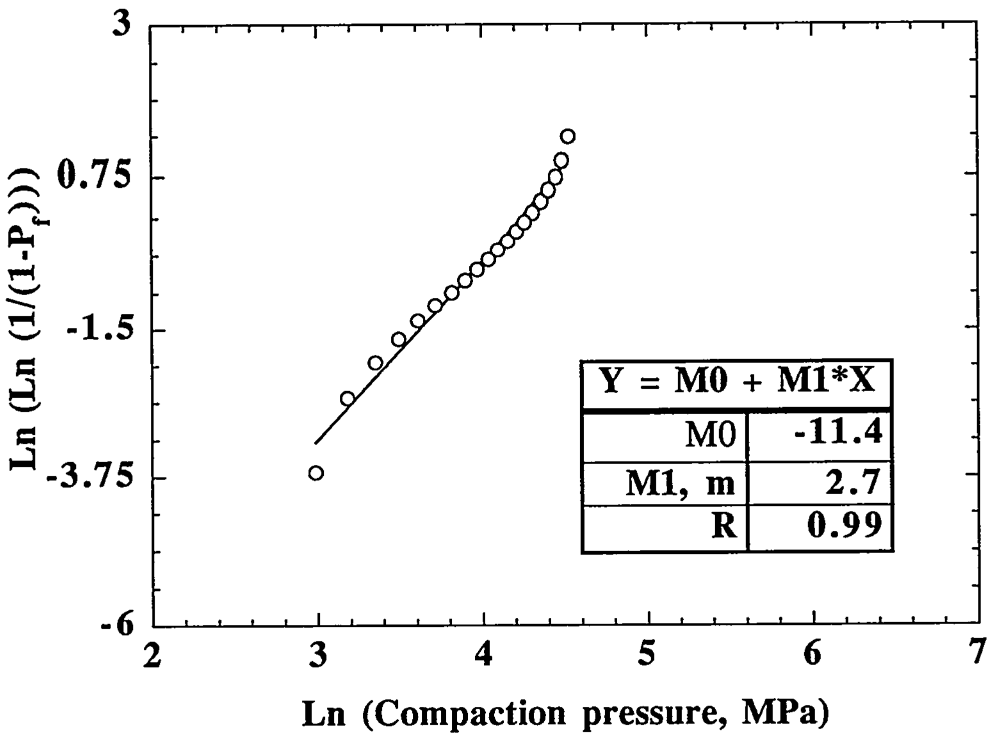

Figure 9. The pressure data were assumed to be equivalent to strength data and were plotted as a Weibull plot. The curve is similar to that obtained using DC strength data, with similar values of m and σ_o.

testing individual granules. Finally, the reasonably successful application of Weibull statistics to analyze granule fracture and granular materials compaction indicates that it may be possible to model compaction using the weak link theory, whereby an assemblage of granules is viewed as the links of a chain, and failure of the weakest granule (i.e., the weakest link) leads to further rearrangement and compaction within the compact.

Acknowledgments

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