A Review of Ceramic Powder Compaction†

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Abstract

The compaction behavior of ceramic powders is reviewed from the viewpoint of the factors involved and how these relate to green microstructure. The basis for the observed linear relationship between the logarithm of compaction pressure and relative density is discussed, and five generic types of compaction behavior are presented. The relation of granule properties to compaction behavior, green strength and green microstructure are discussed. Die friction, springback, die filling and green microstructure evolution are reviewed, and the needs for a better understanding of pressing are discussed.

1. Introduction

Powder processing is the major technique for forming ceramic products, although vapor processes, chemical processes and melt processes are also important classes of ceramic fabrication technology. Of the numerous green forming techniques used in ceramic powder processing, pressing (die or isostatic) is the predominant technique used today because of its low cost, high speed and intermediate shape-forming capability. In recent years, however, little research has been conducted on pressing, while considerable research has been conducted on injection molding, colloidal processing and pressure casting. Currently, there has been a strong increase in the interest in pressing for two reasons. First, many large volume markets for structural ceramics such as automotive valves will only be realized if low cost parts can be manufactured, which probably will require pressing. The second factor is the demand for increased quality for products such as spark plugs that are formed by pressing and have been in production for many years. This will require a higher level of understanding of pressing science and technology.

The quality of the microstructure of a sintered powder compact depends strongly on the quality of the green compact, which in turn is determined by the behavior of powder during compaction. The compaction behavior of a powder is determined by the characteristics of the powder. Understanding the relationships among powder characteristics, compaction behavior, green microstructure, final microstructure and properties is critical to the manufacture of high quality, low cost ceramic products to capitalize on potential markets.

This paper reviews ceramic powder compaction from the viewpoint of the factors involved and how these relate to green microstructure.

2. Basis for Pressure Density Relationship

In 1978 Whittemore(1) reviewed ceramic powder compaction. He cited work by Walker(2) in 1923 that noted a linear relationship between the logarithm of compaction pressure and the relative volume of the compact for several powders. He also cited Balshin's(3) work in 1938 which reported a similar compaction behavior and gave the following relation:

\[ \ln P = AV + B \]  

where \( P \) is the compaction pressure, \( V \) is the volume fraction of the powder in the die and \( A \) and \( B \) are empirical constants. He called \( A \) the pressing modulus and considered it analogous to Young's modulus. Several authors have noted the same relationship for loose(4), precompacted(5) and spray dried(6) powders.

This relationship can be explained by the relationship between the compressive strength and porosity for brittle materials. In 1953, Ryshkewitch(7) showed that the relationship between compressive strength and porosity for alumina and zirconia was given by:

\[ \ln S = \ln S_0 e^{bp} \]  

where \( S \) is the compressive strength and \( p \) is the volume fraction of porosity. Duckworth(8) noted that this relationship could be written as

\[ S = S_0 e^{bp} \]  

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where $S$ and $S_0$ are the compressive strength and the calculated compressive strength of a nonporous body, respectively, and $b$ is an empirical constant. Taking the logarithm of both sides gives

$$\ln S = \ln S_0 - bp$$  

(4)

Substituting (1-V) for $p$, gives

$$\ln S = b V + (\ln S_0 - b)$$  

(5)

If compaction pressure is substituted for compressive strength, this equation has the same form as Equation (1).

In 1959 Knudsen\(^{(9)}\) showed that the logarithm of contact area of ideally packed spheres was proportional to the fractional theoretical density, as an assembly of spheres is densified by deformation at the contacts between spheres. Based on this result, he suggested that the relationship between the logarithm of compressive strength and porosity was a result of the semi-logarithmic relation of critical load bearing area (contact area between spheres) and porosity. If powder compaction is considered as a compressive test of an assembly of powder particles/granules, then it is reasonable to expect a semi-logarithmic relation between relative density and compaction pressure. More recently, Thompson\(^{(10)}\) showed that applied pressure in powder compaction should be related to real contact area between particles in the compact, assuming no breakage of aggregates.

3. Types of Experimental Compaction Behavior

Experimental data show that powder compaction data for ceramic powders often show distinct pressure regimes, each of which shows a different linear relationship between the logarithm of compaction pressure and relative density. In these cases a different mechanism controls compaction in each region. Mort et al.\(^{(11)}\) have reported an automated technique for generating compaction curves. Powders without aggregates that are not spray dried or granulated by other techniques show a linear compaction behavior on a plot of logarithm of pressure versus relative density from 0.1 MPa to 650 MPa\(^{(12)}\) (Figure 1, Curve 1). This case compaction is controlled by interparticle friction, which controls particle rearrangement. Powders containing aggregates that are weak enough to be fractured during compaction and are not granulated show a two-stage compaction curve on a similar plot. The first region is controlled by interparticle friction, and the second region is controlled by the compressive strength of the aggregates (Figure 1, Curve 2)\(^{(4,12)}\). Aggregate fracture occurs at the contact points between aggregates where the local stress is very high and can exceed the compaction pressure by a factor of 100 or more. This is the type of compaction behavior exhibited by calcined ceramic powders that have not been reduced to their ultimate particle size, unless the aggregates are so strong that they do not show appreciable fracture during compaction. A typical metal powder also exhibits this type of compaction behavior, although the second region of the compaction curve is controlled by plastic yielding at the particle contacts\(^{(4)}\) (Figure 1, Curve 3).

Spray dried ceramic powders typically show a three-stage compaction behavior\(^{(6)}\) (Figure 1, Curve 4). Stages I, II, and III are each controlled by a different mechanism. The first stage is controlled by intergranule friction, which does not usually result in any significant compaction. The second stage begins when compaction begins to be controlled by plastic deformation and/or crushing at the granule contact points. This pressure is called the yield pressure\(^{(6)}\). During stage II, the compaction is primarily closing the intergranular pores. The slope of the compaction curve in stage II is the pressing modulus\(^{(13)}\). As compaction continues, the intergranular pores close, and the third stage of compaction begins. In this stage, compaction is controlled by interparticle friction within the granule relics as the intragranular porosity is removed. In this stage the compaction behavior is similar to that of the same powder-binder combination, if it had not been spray dried. A spray dried powder may show another stage which is controlled by fracture of powder aggregates, if the powder has not been reduced to its ultimate particle size and the aggregates are weak enough to show appreciable fracture during compaction (Figure 1, Curve 5). Further analyses of the above relationships are needed in order to improve product quality and manufacturing yields.

For spray dried powders, it is important to remove intergranular pores during pressing, since these pores are generally too large to be removed during sintering. Therefore, parts should be pressed in the stage III region. A transition region between stages II and III occurs because of imperfect packing of granules, a distribution in granule size, a hardness difference among granules or defects in granules such as hollow granules. As a result, intergranular pores close over a range of pressing pressures. As the number of remaining intergranular pores begins to

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Compaction Pressure.

MPa

Fig. 1 Generic shapes of compaction curves.

Curve 1, powder without aggregates that has not been spray dried.

Curve 2, powder containing aggregates which are weak enough to be broken during compaction and has not been spray dried.

Curve 3, metal powder that exhibits plastic deformation at particle-particle contacts.

Curve 4, spray dried powder without aggregates.

Curve 5, spray dried powder that contains aggregates which can be broken during compaction.

decrease, the pressing curve begins to show a transition from stage II to stage III. Unfortunately, isolated intergranular pores can persist well into stage III. This presents a major challenge to the use of pressing as the green forming technique for structural ceramics such as silicon nitride automotive valves, where the cost advantage of pressing over other green forming techniques is critical to market penetration for this and similar components. Furthermore, the demand for increased quality presents similar challenges for products such as spark plugs, that are currently produced by pressing.

Commercial pressing is normally done in the early stage III region. Pressing further into stage III reduces the frequency of residual intergranular pores, but springback becomes more of a problem. Springback increases linearly with compaction pressure and density only increases with the logarithm of pressure. Two other reasons to press early in stage III are, first, that many pressing defects are caused by springback and, second, the capital cost and maintenance costs of presses and dies increase with compaction pressure.

4. Selection of a Binder System

Selection of a binder system for a pressing powder depends on many factors(14). Some of these are:

- Granule plasticity and strength
- Green strength
- Green machinability
- Die friction
- Springback
- Spray drying behavior
- High speed pressing response
- Internal coefficient of friction
- Binder removal/residue

5. Granule Properties

Ideally, spray dried granules should deform plastically to partially fill the intergranular pores and then fracture to assure knitting of particles across the granule-granule interfaces and to reduce the compaction pressure required to close the intergranular pores. Coupelle et al. (15), Baklovti et al. (16) and Nebelung et al. (17) have studied the mechanical behavior of individual spray dried granules during compression between flat platens. Their results indicate that granules deform plastically until the binder system coatings on individual particles are extruded from between particle-particle contact points. The granules then fracture. If granules do not fracture, knitting at the interface generally does not occur. This compromises green strength and leaves low density planes in the green microstructure, especially if an external lubricant is used or if some binder migration occurs during spray drying. Fracture of granules also allows significant particle rearrangement which can reduce problems with residual intergranular pores. However, some amount of plastic deformation of granules is needed for high speed pressing to maintain the high permeability created by the large intergranular pores which allow rapid air removal in the early stages of pressing. Plastic deformation of granules also reduces density gradients caused by particle interlocking, such as that shown in radial compaction of solid or thick-wall cylinders.

The green strength required to allow parts to survive the manufacturing process up to firing is a major factor in binder selection. Some parts such as thin electronic substrates or capacitors require high green strength to avoid mechanical damage. Others such as spark plug blanks require little green strength, but must be compatible with green machining(18). The percentage and type of binder, plasticizer and lubricant in the binder system must be tailored to these and other requirements. Where high green strength is required, a strong binder system is needed. This requires high pressing pressures to reach the stage III region and remove intergranular
Where a low green strength is acceptable, a soft binder system can be used to achieve better compaction properties, since the stage III region can be reached at low pressing pressures. Spray drying slurries often contain a high percentage of solids to minimize the energy required for drying by minimizing the percentage of water to be removed. This can lead to granules with higher densities than what can be achieved in subsequent pressing. This guarantees residual intergranular pores. Several types of granulation other than spray drying have been used or are currently under development. These include drum pelletization, fluid bed granulation and freeze granulation. Pelletization produces granules that are hard due to their density, which makes intergranular pores difficult to eliminate. Mechanically agitated fluid beds are widely used in the pharmaceutical industry and are gaining wider acceptance in the ceramic industry. Nyberg et al. reported that freeze granulation prevents binder migration to the outside of the granules which is often a problem in spray drying. This technique involves spraying a powder slurry into liquid nitrogen followed by freeze drying to remove the solvent. The authors claim that the cost of this process compares favorably with spray drying. This process is also reported to give higher quality, more uniform microstructure than can be achieved with spray dried powders. They conclude that the advantages result from the ability to design the density of granules, since the granule density is fixed by the percentage of solids in the starting slurry. Other advantages of this process are the ability to eliminate binder migration problems and the ease of preparing a granulated powder that is free from defects such as hollow granules, satellites and non-spherical granules, all of which are difficult to eliminate from spray dried powders. Thus, the granule properties can be designed to allow complete removal of intergranular pores during compaction.

6. Precompaction

One commonly used technique for increasing the fill density of a powder is to prepress and granulate the powder. This is a very effective technique for increasing the die fill density of the powder without spray drying. This reduces the compaction ratio (die fill density/pressed density), which helps reduce density gradients in green parts, allows for faster pressing speeds and allows thicker parts to be pressed in a die of a given size. Bruch showed that the final compaction pressure must exceed the prepressing pressure by a factor of about two to eliminate porous regions in the final sintered part. These porous regions are caused by residual pores between the granules of the prepressed and granulated powder. These are analogous to residual intergranular pores in compacts pressed from spray dried powders and are generally too large to be removed during sintering.

7. Die Friction

Die friction is a key problem in die pressing of ceramic powders and is the major cause of density gradients in die-pressed parts. In combination with springback, it is also the cause of many of the common defects in die pressed parts. Studied die friction and concluded that slippage occurs within the powder (powder failure stress) rather than between the powder and the die wall. However, for carbide dies with a smooth surface finish and a powder containing a good binder-lubricant system, slippage probably occurs between the powder and the die wall, at least at low pressures. As the surface finish of the die deteriorates due to wear, slippage between the powder and the die wall becomes less likely.

Thompson showed that end capping originates from the restraint of elastic springback when the punch pressure is released. He considered three terms: length/diameter ratio, powder fluidity index and the coefficient of friction at the die wall. The powder fluidity index is the ratio of the radial/axial stress at the die wall. For a fluid, this ratio is one. For ceramic powders, it depends on the internal coefficient of friction of the powder/binder system. Thompson concluded that addition of an internal lubricant to the powder may increase or decrease the die wall friction force. This results from the increase of the powder fluidity index as the internal coefficient of friction is decreased through the addition of a lubricant. Therefore, the increase in the radial force may more than offset the reduction in the friction coefficient. Another technique for reducing end capping and other defects caused by springback is to incorporate a hold-down pressure in the pressing cycle. This involves maintaining a large percentage of the pressing pressure on the part during ejection from the die. Modern presses are designed to provide a hold-down pressure.
External lubricants are often added to spray dried powders to reduce die friction. Internal lubricants are added to the slurry before spray drying and are distributed on the surfaces of the powder particles throughout the granules. However, external lubricants such as stearates are added to the powder after spray drying and coat only the external surfaces of the granules. Since the surface area of the granules is only a fraction of the surface area of the powder, a very small percentage of lubricant is needed to significantly reduce die wall friction. Only the lubricant on the granule surfaces that contact the die wall is effective. However, it has the same effect as lubricating the die wall between each pressing, which is not possible in high speed pressing. Often, if the internal binder system contains enough lubricant to produce the low die wall friction desired, the binder system requires such a high percentage of lubricant that pressing speed must be compromised due to die sticking problems.

8. Springback

Springback in dry pressing is a result of stored elastic energy in the part at the compaction pressure and may also result from trapped air. Differential springback within a part must also be considered, since it can result in pressing defects. The stored energy is usually the sum of several components. The contributing components can be separated into those caused by the organic components and those caused by the inorganic components. The organic components typically have both an elastic and a visco-plastic (time-dependent plastic) component. The contribution of the organics to total springback may be the dominant factor, since the elastic modulus of the organics is considerably less than that of the inorganic components. However, the elastic strain stored in the organic components may not be fully recovered, since the recovery force may not be high enough to overcome the constraining force caused by interlocking of the inorganic particles and their adhesion from the binders or other forces. Since the binders may make a major contribution to springback, the binder system is designed to have a glass transition temperature below the pressing temperature. This makes the binder behave as a liquid rather than a solid and substantially reduces springback caused by the organic components. However, springback still increases with the percentage of organics in the powder. For high speed pressing, it is important to recognize that the glass transition temperature of a polymer is strain-rate dependent. If sufficient time is not allowed for stress relaxation by viscous flow at the pressing pressure, the binder will behave as an elastic solid rather than a viscous liquid and significantly increase springback.

The springback, due to elastic energy stored in the inorganic constituents, can be minimized by using spherical particles with a high elastic modulus. Sharp corners, non-equiaxed particles and porous aggregates all increase springback. Every particle can be viewed as a miniature spring. Contact points in the compact that involve sharp corners experience a higher stress and, therefore, a higher stored elastic strain than spherical contacts. Another reason powders with sharp corners exhibit high springback is that they increase particle interlocking, and interlocking particles can act like porous aggregates. Porous aggregates increase springback for two reasons. First, they act like particles with a significantly lower elastic modulus than dense particles, because the bridges at the particle-particle contacts in the aggregate may have a cross-sectional area that is only a fraction of that of the particles themselves. The second reason is that they store a significant strain in bending. This becomes much more significant as the shape deviates from equiaxed. Platy particles such as clay or talc, fibrous particles and non-equiaxed particles are also major contributors to springback, since these shapes can store a large amount of strain in bending.

As compaction begins, the powder contains considerable void space filled with air. The air must either escape from the die or be compressed, and this must be taken into account in designing the pressing cycle. Powder permeability, die clearance, green-body size, pressing speed/cycle and binder type/percentage all affect air removal that can cause defects during removal or on pressure release. Some dies are designed with vacuum ports to allow faster pressing without encountering defects due to trapped air. In designing the pressing cycle, it should be remembered that air removal is a function of volume reduction and not pressure. Therefore, air removal considerations are critical in the low pressure portion of the pressing cycle. Many pressing cycles are designed with one or two pressure relief cycles to enhance air removal.

9. Die Filling

Die filling is a critical part of a pressing cycle. Once the die is filled, the final compact microstructure
is relatively fixed. High speed pressing cycles usually include withdrawal of the bottom punch as the die is filling to speed filling and to avoid air pockets or air channels during the die fill. Since most cycles fill by volume rather than weight, it is critical to reproduce the fill density from part to part and from point to point throughout the die. This requires a reproducible granule density and granule size distribution in the spray dried powder. Even with a uniform spray dried powder, any size segregation in the powder during delivery to the press or in die filling will result in point-to-point or part-to-part variation in fill density and lead to density variation in the pressed part. Modern presses are designed to eliminate size segregation, since spray dried powders are very susceptible to this problem.

10. **Green Microstructure Evolution**

   In order to understand how the microstructure of a powder compact develops during pressing, fracture and as-pressed surfaces can be examined using a scanning electron microscope after pressing at a sequence of pressures. These microstructures can be used along with the compaction curve, powder characteristics, the microstructure of the sintered sample and mechanical properties of the material to better understand how to fabricate a quality material. **Figures 2 and 3** show the pressed surfaces and fracture surfaces, respectively, for a high alumina porcelain composition which was spray dried by the manufacturer and pressed at several pressures. The pressed surfaces in **Figure 2** show the progressive deformation and fracture of granules as the compaction pressure is increased. It also shows that isolated intergranular pores persist to very high pressures. Uematsu et al.\(^{28}\) developed an excellent technique for direct observation of internal voids in green bodies. They infiltrated green bodies with a fluid that has a small mismatch in index of refraction with the powder. This makes the green body almost transparent, but voids throughout the body are visible. The intergranular pores are exaggerated in **Figure 2** by the fact that the boundary conditions are different adjacent to the die plunger than inside the compact. However, the wide variation of intergranular pore closure pressures is clearly shown. **Figure 3** shows fracture surfaces from compacts of the same powder. These clearly show the deformation and fracture of the granules. Although it is not possible to tell whether the granules fractured on pressing or on fracture, whole granules tend to pull out of the fracture surface if they did not fracture and knit across intergranular boundaries during compaction.

11. **Conclusions**

   Much is known about ceramic powder compaction, but much more needs to be understood if it is to be used to produce the high quality, low cost ceramic products that are needed to develop markets for new products and provide the improved quality demanded for current products.

   Many defects in the final microstructure can be traced to the green microstructure. Residual pores caused by hard or defective spray dried granules or by intergranular pores due to incomplete compaction in
isolated locations in the part are a problem. These pores are generally too large to be removed during sintering and cause defects in the final microstructure. More needs to be known about the origin of these pores and how to avoid them.

Another key problem is density variation within parts and among parts, since this affects both part quality and the ability to achieve tight dimensional tolerances. Die wall friction is a key reason for density variations, but the powder fluidity index is also important, since a low internal coefficient of friction helps reduce powder bridging that also causes density variation. Improved lubricants are needed to reduce the coefficient of friction both at the die wall and within the powder. A better quantitative understanding of the factors that affect differential shrinkage is also needed, since dimensional tolerances are also affected by differential shrinkage.

Springback is another area where a better understanding is needed. A quantitative understanding of the factors that affect springback is needed, since the combination of springback, die friction, green strength and the pressing cycle all interact to dictate defects in pressed parts. One factor that has not been given enough attention is the visco-plastic or time-dependent plastic properties of organic additives. The visco-plastic properties of an organic additive may cause the binder to behave very differently in high speed pressing where strain rates become very high and relaxation times very short compared to laboratory pressing trials.

Much can be learned by application of finite element analysis to powder compaction. With the computing capability now available, finite element analysis integrated with a strong experimental compaction program should lead to the major advances in the quantitative understanding of powder compaction that are needed to improve the quality of ceramic components formed by powder compaction.

Bibliography

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20) Takahashi, H., Shinozara, N., Okumiya, M., Uematsu, K., Junichiro, T., Iwamoto, Y. and Kamiya H., “In-

Author’s short biography

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Dale Niesz obtained a bachelor of science (cum laude), a master of science, and a doctor of philosophy degrees in Ceramic Engineering from The Ohio State University in 1962, 1963 and 1965, respectively. He spent 22 years in contract research at Battelle Columbus Laboratories, where he served as a research engineer, manager of ceramic research and manager of materials research. He joined Rutgers, The State University of New Jersey, in 1987 as a Professor in the Department of Ceramic Engineering. He currently serves as Director of the Center for Ceramic Research and Chair of the Department of Ceramic Engineering.

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