COMPRESSION AND COMPACTION CHARACTERISTICS OF SELECTED FOOD POWDERS

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I. INTRODUCTION

Before exploring the different findings in the modeling of unpacked powders behavior under pressure or vibration modes, and in order to understand the different microstructural descriptions of compression and compaction phenomena encountered during different food powder processing operations, it is important to revisit some definitions for the fundamental descriptors used

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in the mechanics of materials and food powder physical properties studies. Depending on the bibliographical source revised, some of these definitions may lack in accuracy or may vary according to the approach received for their determination.

Compression and compaction parameters such as density, porosity, stress, and strain are introduced. Furthermore, other factors of importance in stress measurement of powder beds in different situations such as compression, tension, shearing, and impact are explained. The fluidity of food powders is a controversial topic in food powder characterization and of top relevance in the food industry. Concepts such as compressibility and other flowability indicators are important for understanding the effects of compression phenomena during storage, production, and handling. Food powder strength concepts such as hardness relate to other concepts like attrition, which are a common occurrence in compaction processes.

We also describe testing for compression evaluation using the Brazilian test, uniaxial compression test, cubical triaxial tester, high hydrostatic pressure (HHP) method, unconfined yield stress test, impact and shear tests, as well as vibration methods for compaction-extent follow-ups. The interparticle/intraparticle adhesive forces that naturally exist in powders in fine static powder beds and agglomerates and that intrinsically rule compression and compaction phenomena are portrayed. This introduction also depicts compression mechanisms that occur during testing and in different unit operations in fine particles and agglomerates, along with typical compression curves and other compaction mechanisms such as attrition and segregation.

A. COMPRESSION AND COMPACTION-RELATED PROPERTIES

The terms *compression* and *compaction* are related to the ability of a loose powder to form a powdered compact by decreasing its initial volume. Whereas *compression* matches the idea of die pressing of powders, *compaction* describes the kinetic or vibratory rearrangement of particles within a certain bulk structure. Properties such as porosity, bulk density, compressibility, pore size distribution, instant properties, and flowability are closely related and play a significant role in the evaluation of processing, handling, and storage conditions (Barbosa-Cánovas *et al.*, 1987; Moreyra and Peleg, 1980).

1. Powder density and bulk porosity

Density (ρ) is defined as the unit mass per unit volume measured in kilograms per squared meter in SI units and is of fundamental use for material property studies and industrial processes in adjusting storage, processing,

packaging, and distribution conditions. In particular, bulk density is one of the properties used to specify the end product derived from grinding or drying. There are three main types of density: true density, apparent or particle density, and bulk density. A newer concept for powder density has also been introduced as "ultimate bulk density" and has been identified as "Barbosa-Cánovas" density (Barbosa-Cánovas and Juliano, 2005; Yan *et al.*, 2001). It is recommended that before using or comparing density values from the literature, one should verify the density determination method used because other authors have used different names for the same type of density.

a. True particle density (ρ_s). Also denominated as *substance density,* the true particle density represents the mass of the particle divided by its volume excluding open and closed pores, that is, the density of the solid material composing the particle. In this case, to measure the powder volume, the substance is broken, milled, or mashed to guarantee that no external or internal pores remain. Some metallic powders can present true densities at around 7000 kg/m^3, whereas most food particles have considerably lower true densities at 1000–1500 kg/m^3.

b. Apparent particle density (ρ_p). This is the mass over the volume of a sample that has not been structurally modified. Volume includes internal pores not externally connected to the surrounding atmosphere and excludes only the open pores. It is generally measured by gas or liquid displacement methods such as liquid or air pyknometry.

c. Bulk density (ρ_b). This is measured to include the volume of the solid and liquid materials, as well as all pores closed or open to the surrounding atmosphere. Powders have "loose bulk density" or measured density after a powder is freely poured into a container, and they have "compact density" after it is allowed to compress by mechanical pressure, vibration, and impact (Peleg, 2004). Another type of density more related to the compaction of powders is "tap density," which is the density of a certain powder mass over the resulting volume of powders after being tapped or vibrated under specific conditions. In particular, the "ultimate bulk density" (Yan *et al.*, 2001) is the constant density reached after compressing an agglomerated powder over a critical high-pressure value, wherein no open or closed pores remain.

d. Porosity (ϵ). The volume fraction of air (or void space) over the total bed volume is indicated by porosity or voidage of the powder. Based on given distinctions among densities and considering air density as ρ_a, the definition of bulk density is

$$\rho_b = \rho_s(1 - \epsilon) + \rho_a\epsilon \tag{1}$$

Because air density is small relative to powder density, it can be neglected. Porosity can thus be calculated from Equation 2, excluding the air pores within the volume of bulk mass:

$$\epsilon = \frac{(\rho_s - \rho_b)}{\rho_s} \tag{2}$$

2. *Strain during compression*

The relative deformation or dimension change due to force in the size or shape of a body, with respect to its original size or shape, defines the strain in a certain material. Strain is a measure of the deviation or displacement of the components (molecules, atoms, ions) throughout the material from their normal position. Strain can be linear (changing with tensile or compressive forces in a longitudinal dimension) or shear (angular changes due to force between two lines). Strain can be expressed depending on the direct observable longitudinal change in a body or change in gauge length ΔL from strain to voltage conversion (American Society for Testing and Materials [ASTM], 1986). Engineering strain $\in_{\text{Engr}}$ directly expresses the change in deformation with respect to an initial length $L_{0:}$

$$\in_{Engr} = \frac{\Delta L}{L_s} \tag{3}$$

On the other hand, natural strain (also called *logarithmic* or Hencky's strain) $\in_{\text{True}}$ results from integration of infinitesimal strains (Swyngedau *et al.*, 1991). However, it is only applicable to particulate materials forming a stable or cohesive structure.

$$\in_{True} = \ln\left(\frac{1}{1 - \in_{Engr}}\right) \tag{4}$$

3. *Stresses during compression*

Stress, or force per unit area (SI units Pa or N/m^2), has been defined as the intensity of the internal components of forces in a certain point through a given plane of a body. *Compressive stress* (or pressure) refers to the perpendicular components toward a normal plane on which compressive forces act. Different denominations can be used for stresses that characterize compression of a certain volume of powder mass: natural and engineering stress, compressive, tensile, or shear stress, yield stress, unconfined yield stress, and principal stresses.

a. Engineering, natural, and confined stress. *Engineering stress* σ_{Engr} simply refers to the ratio of the force applied over its initial area of compressed or stretched solid mass. On the other hand, natural stress, also called *true stress,* is the axial stress in a tension or compression test calculated on the basis of the instantaneous cross-sectional area instead of the original area (Mao *et al.*, 2000; Mohsenin, 1986).

$$\sigma_{True} = \sigma_{Engr}(1 - \in_{Engr}) \tag{5}$$

As in true strain, the expression above takes into account cross-sectional area changes in a certain cohesive structure (or cake) of powdered material. If the material is isotropic, another possible expression that includes the Poisson's ratio μ (the ratio of transverse strain and axial strain resulting from uniformly distributed axial radial stress during static compression of the material in absolute value). The Poisson's ratio, or bulk modulus, permits prediction of the transverse contraction or expansion that occurs when a stress is applied longitudinally.

$$\sigma_{True} = \frac{\sigma_{Engr}}{(1 - \mu \in_{Engr})^2} \tag{6}$$

Because easy-flowing powders cannot remain piled as an unconfined structure, allowing small strain changes within, true stress (Equation 6) cannot be used to characterize compression. Thus, the concept of *confined uniaxial compression stress* has been introduced to help characterize compressive stress in powders. Confined uniaxial compression stress is the force exerted by a piston that compresses a certain powder over the piston area. The powder is generally poured and confined in a container that fits closely to a piston's wall.

b. Compressive, tensile, and shear stress. Compared to compressive stress, tensile stress refers to the normal stress due to forces directed away from the plane. Numerous experimental results have shown there is no simple correlation between tensile and compressive strength, although the ratio depends critically on the geometry of the specimen and the amount of plastic yielding at the point of load application (Bika *et al.*, 2001).

Tensile stress is used to characterize cohesiveness between particles, or in a certain powder cake, coating resistance in an encapsulated powder. Shear stress refers to the stress component tangential to the plane on which forces act and is mainly used to determine frictional properties (e.g., angle of internal friction) between particles under a pressure load. Furthermore, because individual particles predominantly slide across each other in a shearing action during flow, shear stress measurement allows determination of flow properties.

c. Elastic and ductile stress regions yield stress. In a stress–strain or force–deformation curve (Figure 1), a material experiences elastic deformation when it returns to its original shape once the force is removed. If the stress exceeds the elastic limit, the material undergoes permanent (inelastic) deformation until it reaches the yield stress point when it begins to flow (region of ductility). The yield point is defined as the first stress in a material and is less than the maximum attainable stress, at which point an increase in strain occurs without an increase in stress (ASTM, 1986). After the yield point, applied stress will act until the material finally breaks. This process defines the elastic stress limit, yield stress, the region of ductility, and breaking stress represented in Figure 1. A food powder may be hard or soft; increased hardness is correlated with an increase in the modulus of elasticity (or slope in the linear elastic region).

A brittle material breaks soon after the stress exceeds the yield stress. Brittleness is a measure of the size of the region of ductility. Conversely, a ductile material can deform considerably without breaking. Another property is toughness. A tough material has the ability to resist the propagation of cracks. Fibers impart toughness by relieving stress concentrations at the end of the cracks. The opposite of toughness is fragility. The ultimate stress in Figure 1 is called *breaking stress,* which describes the particle breakage that occurs along cracks or defects in the structure. A coarse particle with many defects can be broken under small stress with very little deformation.

d. Impact stress. Impact stress results from collision forces that are rapidly applied to the material to produce stress waves that instantaneously leave the region of contact when force is removed. Particles can respond

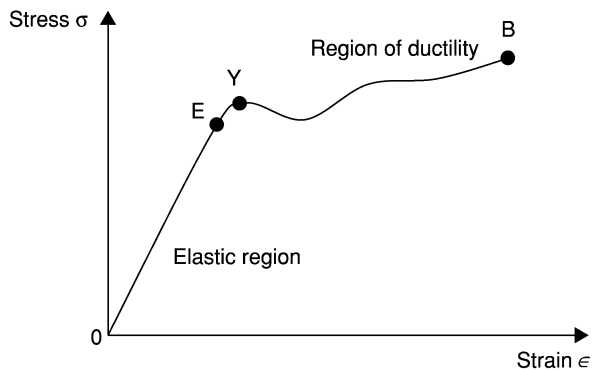

FIG. 1 Stress-strain diagram for various types of solids. B, breaking point; E, elastic limit; EY, elastic deformation; OE, elastic region; Y, yield point; YB, region of ductility (adapted from Loncin and Merson, 1979).

elastically without residual deformation or plastically depending on the dynamic yield pressure of the material. When plastic impact occurs, the kinetic energy is converted into permanent deformation of the material and eventual dissipation of this energy in the form of heat. Impact stress is useful to indicate powder or capsule mechanical damage during handling operations.

e. Unconfined yield stress. The unconfined yield stress f_c indicates the maximum compressive stress a cohesive particle array is capable of sustaining at a particular porosity (Mohsenin, 1986; Peleg, 1978; Schubert, 1987). It also represents the strength of the cohesive material at the surface of an arch (Figure 2), which resists a lower stress induced by its own weight (Teunou *et al.*, 1999). In flowability characterization, the unconfined yield stress refers to a situation in which the physical configuration of the system allows the powder to flow before massive comminution of the particles occurs.

f. Principal stresses. The general state of stress at any stressed point is defined by three orthogonal planes, on which there are zero shear stresses (Lambe and Whitman, 1969), which are called *principal planes*. The normal stresses that act on these three planes are called the *principal stresses*. The largest of these stresses is called *major principal stress* σ_1, the lowest minor principal stress σ_3, and the third intermediate principal stress σ_2 (Sandor, 1998).

4. Compressibility

In general terms, *compressibility* refers to the variation in bulk density with respect to consolidating confined pressure acting on a powder bed. Bulk density (in terms of apparent, compact, or tap density) and normal stress have been associated in empirical logarithmic or semilogarithmic relationships, from which a constant slope value is defined as *mechanical compressibility*. Simultaneous decrease in a powder's loose bulk density and increase in compressibility indicate greater attractive and cohesive interactions among powders.

In this chapter, we address in considerable depth empirical methods and models found in the literature, as well as variables that affect compressibility such as moisture, temperature, and particle size and shape. Compressibility can be used in feeder designs to calculate loads that act on a feeder or gate and angle of wall friction to calculate the pressures acting perpendicular to the hopper wall. Furthermore, it can be used for quality control to determine the resistance of materials to breakage, from the production process to the consumer.

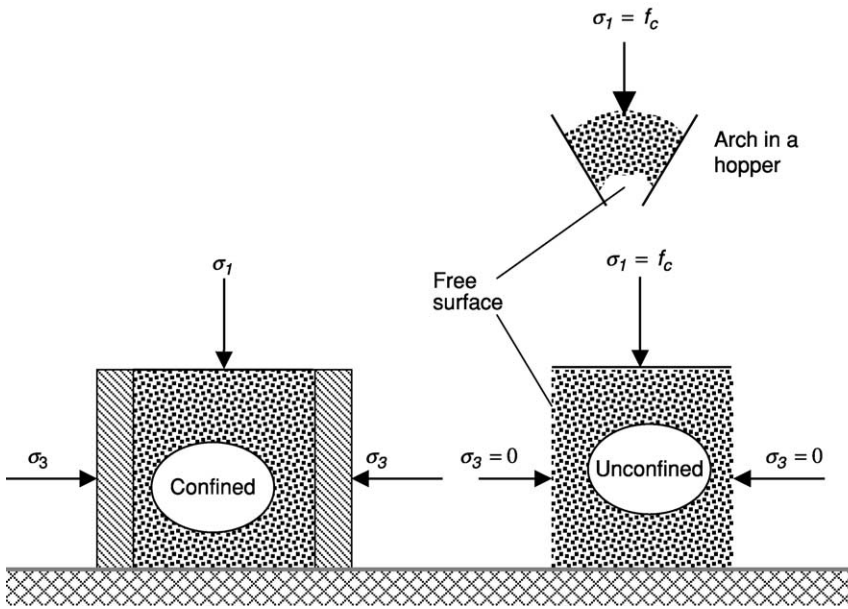

FIG. 2 The free surface of a powder under consolidation represents the conditions of the minor Mohr circle. Under minor principal stress $\sigma_3 = 0$; the major principal stress σ_1 is defined as the unconfined yield strength f_c and represents the strength of the powder at the free surface of the arch (adapted from Bell, 2001).

5. *Fluidity and flow characterization*

The fluidity of a powder is the ease of flow. It relates to changes in mutual position of individual particles forming the powder bed and depends on frictional and cohesive forces. The dynamic behavior of powder seems to be determined by interparticle forces and packing structure. Because different types of flow occur in industrial processes, this concept is applied only to powder flow processes, where compaction and compression occur. Compaction is mainly related to mechanically forced and vibrating flow (e.g., tumbler and ribbon mixing, vibrating feeding and conveying, and packing), whereas compression is more related to powder briquetting and tableting (e.g., cereal or candy bar production and encapsulation process) or gravitational flow (hopper discharge or packing under particle load).

Within the concept of fluidity is flowability, that is, the ease at which a powder flows through a chute or hopper. Compressive and compaction behavior of powders is important in evaluating flowability, because methods

to determine flow properties like the angle of repose and angle of internal friction account for compressive and compacting mechanisms.

Powders can be classified in two types when referring to powder flowability: noncohesive powders and cohesive powders. Noncohesive (or "free-flowing") powders are those powders in which interparticulate forces are negligible. Most powders are considered noncohesive only when dry and when particle size is more than 100–200 μm (Peleg, 1978; Teunou *et al.*, 1999); finer powders are susceptible to cohesion, and flowability is more difficult (Adhikari *et al.*, 2001).

However, in cohesive powders interparticle forces play a significant role in the mechanical behavior of the powder bed (i.e., their attractive interparticle forces are significantly high relative to the particle's own weight). Therefore, after compressing an open bed of cohesive materials, the bed would likely remain supported only by interparticle forces (Peleg, 1983).

Cohesive powders when poured from a beaker will flow like liquid, but under these conditions, the material has no cohesive strength. If the powder is squeezed against the bottom of the beaker, the material may gain enough strength to retain its shape once pressure is removed. A similar phenomenon occurs inside bins, hoppers, and containers, leading to the formation of arches or ratholes (Carson and Pittenger, 1998). Consolidation pressures range from zero at the surface (Figure 2) to relatively large values at increasing depth within the container where cohesive strength is higher.

A standard for cohesive strength determination is the ASTM D 6128 (or the Jenike method), in which consolidating conditions found in the depth of a bin are reproduced in a shear cell. The shear cell is used to determine the asymptotic shear force (or yield point) a powder can undergo under a predetermined compression load and preshearing. A plot (*yield locus*) of asymptotic shear stress (shear force divided by the cell's cross-sectional area) versus the corresponding normal consolidating stress gives a curve where two parameters can be obtained (Peleg, 1978): *cohesion* C and *angle of internal friction* ϕ. Furthermore, the standard indicates the use of two Mohr circles tangent to the yield locus, which allows determination of two other parameters used for flow property characterization: *unconfined yield stress* and *major principal stress*. Because these properties are important for characterizing the flow of material (e.g., from a hopper), they have been compared with compression properties to determine a more direct and easier method for flowability determination. Some brief definitions of flow properties as compared with compressibility are presented next:

a. Cohesion C. This property is a measure of the attraction between particles and is due to the effect of internal forces within the bulk, which tend to prevent planar sliding of one particle surface on another. Cohesion value is at the intersection of the yield locus with the shear stress axis. Cohesion has been proven proportional to tensile stress in certain powders (Peleg, 1978).

b. Angle of internal friction. This property is a measure of the interaction between particles and is calculated from the slope of the yield locus. In free-flowing powders, it represents the friction between particles when flowing against each other. Therefore, it depends on their size, shape, roughness, and hardness.

c. Flow function. The flow function FF is a complex function that provides a measure of the strength of the cohesive material in the surface of an arch as a function of the stress by which the arch was formed. Schubert (1987) defined it as the ratio of unconfined yield stress and major principal stress. These values are directly obtained from geometrical calculations using the Mohr circles tangent to the yield locus.

d. Angle of repose. The *static angle of repose* is defined as the angle at which a material will rest on a stationary heap; it is the angle θ formed by the heap slope and the horizontal when the powder is dropped on a platform. According to Carr (1976), angles up to 35 degrees indicate free flowability; 35–45 degrees, some cohesiveness, 45–55 degrees, cohesiveness (loss of free flowability); and 55 degrees or more, very high cohesiveness; therefore, there is very limited (or none) flowability. The angle of repose method can roughly indicate flow in small quantities of consolidated powders. It is the actual flowability measurement applied by some laboratories in the food industry for quality control.

6. Hardness

The hardness of powders or granules is the degree of resistance of the surface of a particle to penetration by another body. It is related to the yield stress, considering the characteristics of the uniaxial stress–strain curve for several types of material failures (i.e., transition between elastic and plastic strains). Hardness can be determined as a qualitative property by using the Mohs hardness scale (Carr, 1976). In this scale, 10 selected minerals are listed in order of increasing hardness, by indicating qualitative resistance to plastic flow, so that a material with a given Mohs number cannot scratch any substance with a higher number but will scratch those with lower numbers. Materials different from those included in the scale are referred to as having an equivalent number of hardness as the 10 listed. Quantitative determination can be done through nanotesting and atomic force microscopy by

providing detailed characterization of surfaces of even micrometer-sized particles (Scherge and Gorb, 2001).

A particle property related to hardness is the crushing strength (ASTM, 1986), which refers to the force required to crush a mass of dry powder, or conversely, the resistance of a mass of dry powder to withstand collapse from external compressive load. Crushing strength is the resistance of a solid to compression, a property of paramount importance not just for tablets and capsules. Bulk crushing strength can be evaluated by measuring either the amount of fines produced after compression of a fixed volume of particles at a predetermined pressure or the pressure required for producing a predetermined amount of fines.

7. Attrition

Attrition is a deleterious particle breakdown, which increases the number of particles and reduces particle size, thus affecting particle size distribution. Except for particle size reduction during comminution or grinding processes, attrition is undesirable in most processes. In fact, it is one of the most pressing problems for a wide range of processing industries dealing with particulate solids. In food powders, attrition occurs more frequently in agglomerates, mainly because of their multiparticulate structure. Many food agglomerates possess brittle characteristics that make the product susceptible to vibrational, compressive, shear, or even convective forces received by the particles during processing.

B. COMPRESSION AND COMPACTION EVALUATION IN FOOD POWDERS

In powder technology, great attention has been paid to the general behavior of powders under compression stress. Compression and compaction tests have been widely used in pharmaceutics, ceramics metallurgy, and civil engineering, as well as in the food powder field by researchers to study the mechanisms of particle interactions and to evaluate particles at a bulk level.

Methods to determine compression behavior can be either static (dead load) or based on constantly increasing compression. Several methods for the determination of volume-reduction mechanisms due to compression or compaction have been presented in the literature; the most relevant ones are described in the following sections. Compression mechanisms can be approached from different tests such as the Brazilian test, uniaxial confined compression test, cubical triaxial tester, HHP method, and the unconfined

yield stress. Other compaction properties such as hardness, crushing strength, and attrition tendency can be determined through the use of shear cells or from impact and vibrational tests.

1. Brazilian test

One conventional method is the so-called "Brazilian test," as shown in Figure 3, which is applicable for single particle measurement. In this test, the particle is crushed between two flat rigid platens and the load required to cause fracture is recorded. This commonly used method is usually employed to assess the breakage behavior of brittle particles (Shipway and Hutchings, 1993). It is time consuming because a large number of particles must be measured and there are always wide variations in the fracture load measurements (Adams *et al.*, 1994; Nuebel and Peleg, 1994). Because of these disadvantages, an alternative and more convenient compression test is preferred (Barbosa-Cánovas and Yan, 2003), which is usually called the *confined uniaxial compression test,* as shown in Figure 4.

2. Universal testing machines: Tests for confined uniaxial compression

Universal testing machines (UTMs) allow computer readout and analysis of force-time plots. Special software displays graphs for maximum normal and shear forces during compression tests, as well as creep and stress relaxation curves. Mechanical compressibility and breaking load under tension can also be determined from stress-strain data by using UTMs.

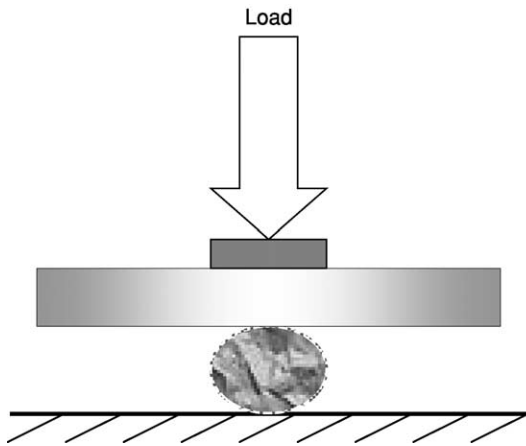

FIG. 3 Brazilian test for a single particle (from Yan and Barbosa-Cánovas, 2000).

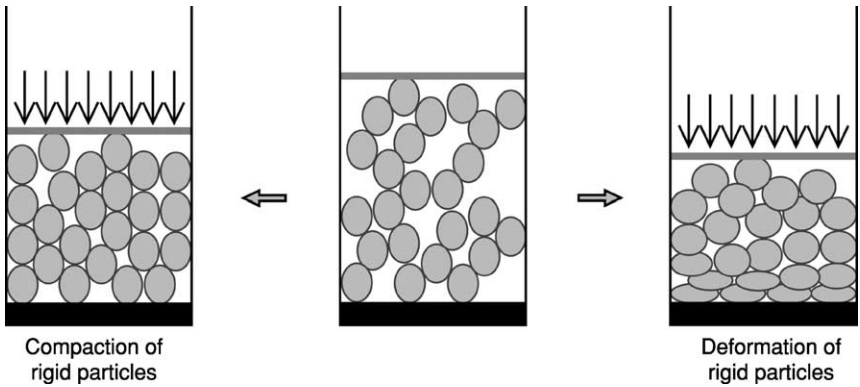

FIG. 4 Confined uniaxial compression test. Bed compression of rigid and deformable particles (adapted from Lu *et al.*, 2001).

The confined uniaxial compression test using a UTM (Figure 5) involves confining a bed of powder in a cylindrical cell and measuring the force applied to the flat-based piston, which is in contact with the top surface of the bed, as a function of the piston displacement. The mass of the bed is constant and the bed height is continuously monitored on a recorder chart. The powder bed is moved by the piston downwards at constant speed. The changes in bulk density, or porosity, of the powder bed versus the compression load are usually expressed by mathematical functions described next. During the compression test the displacement of the piston and the consolidating force are recorded. Using dimensions of the compression cell and the mass of the sample, the actual readings in vertical displacement can be transformed into bulk density by

$$\rho_{bulk} = \frac{m_s}{A(h_0 - x)}, \tag{7}$$

where m_s is the mass of the sample, A is the surface area of the piston, h_0 is the initial filling height, and x is the displacement of the piston.

Different vertical loads can be applied to a bulk solid sample of known mass, and compression of the sample is recorded electronically (Thomson, 1997). With these data, powder contact volume versus compressive force or stress can also be represented. Bulk density of a solid is a function of consolidation stress and changes during flow as the stress changes. Because the mass consolidating load and volume are known, the relationship can be plotted as shown in Figure 6.

This method can successfully evaluate particle attrition (Bemrose and Bridgwater, 1987), flowability (Peleg, 1978; Schubert, 1987), compressibility

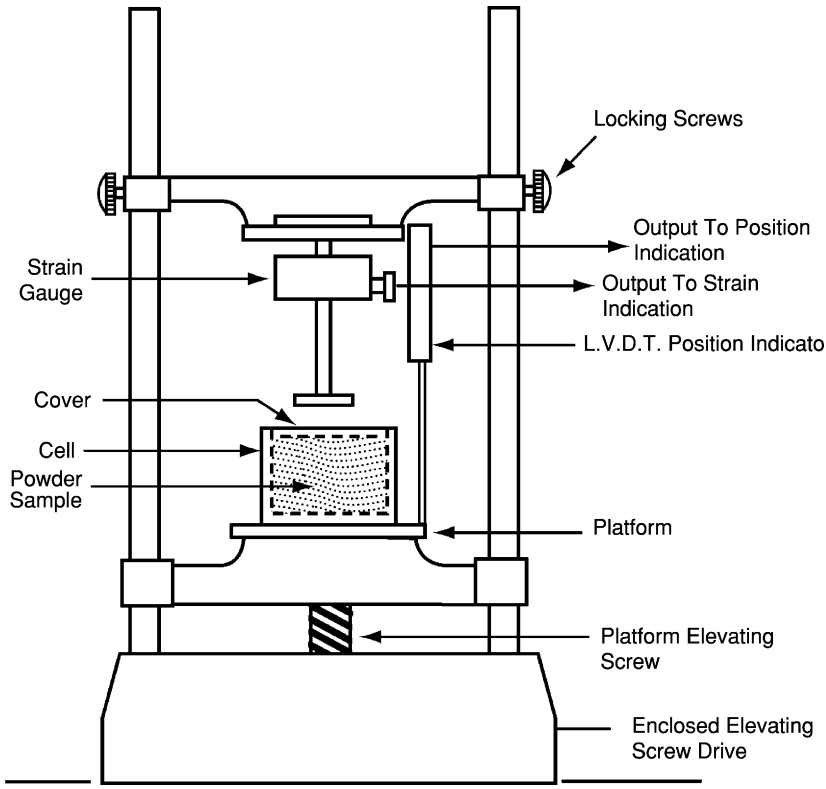

FIG. 5 Diagram example of a universal testing machine for bulk density testing during compression (from Thomson, 1997). LVDT, linear variable displacement transducer.

(Barbosa-Cánovas *et al*., 1987; Malavé-López *et al*., 1985; Yan and Barbosa-Cánovas, 1997), and agglomerate strength (Adams *et al*., 1994). In particular, any agglomerate measurement will be affected by both the breakage properties of individual particles and the deformability of their assembly as a whole (Nuebel and Peleg, 1994). Equipment models used in different research projects are listed in Table I.

One disadvantage that may be encountered during bulk compression is that the geometry of the bed and the powder filling method may have an important influence on results (Nuebel and Peleg, 1994; Yan and Barbosa-Cánovas, 2000). To minimize the wall effects, the cylindrical cell used for the compression test should have a larger diameter with respect to its height. In an interlaboratory study (Cost 90bis Collaboratory Study), Ehlermann and

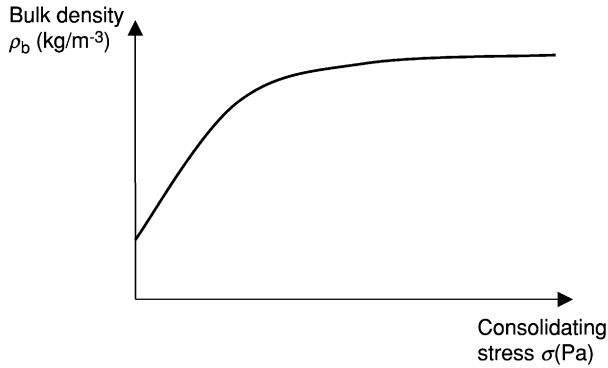

FIG. 6 Typical plot of bulk density versus consolidating stress.

Schubert (1987) proved that friction between the piston and compression cell wall was the main cause of inaccurate results in previous works on compressibility. The use of strain gauges on the inner part of the piston to measure the pressure directly at the powder surface facilitated separating the effects of wall friction from those of true pressure on the surface of the consolidated powder (Figure 7).

3. *Cubical triaxial tester*

A flexible boundary cubical triaxial test is another commonly used test for compression studies (Kamath *et al.*, 1993; Li and Puri, 1996). A picture of a triaxial compression tester is shown in Figure 8. It allows not only the application of the three principal stresses independently, but also constant monitoring of the volumetric deformation and deformations in three principal directions. In a triaxial compression test, the specimen is at an initial isotropic state of stress; then the three pressure lines apply the same pressure at the same rate to all six faces; thus pressure is the same in all three directions (i.e., $\sigma_1 = \sigma_2 = \sigma_3$).

Research has shown that the cubical triaxial tester is useful for investigating anisotropy of cohesive and noncohesive powders and the effect of particle shape and sample deposition methods. For anisotropic materials such as wheat flour, the strains in three principal directions are statistically different (Li and Puri, 1996). Wheat flour was used for the calibration of the triaxial tester to determine parameters related to a hopper design (Kamath, 1996).

TABLE I
SOME EQUIPMENT USED FOR COMPRESSION TESTS

Brand	Model	Material	Author
Instron Universal Testing Machine	1000	Spray-dried coffee	Nuebel and Peleg, 1994
Instron Universal Testing Machine	TM	Instant coffee Wheat flour Rye flour Corn starch Soy protein Malic acid Granular sucrose Ground roasted coffee	Barbosa-Cánovas *et al*., 1987; Hollenbach *et al*., 1982; Malavé-López *et al*., 1985; Molina *et al*., 1990; Moreyra and Peleg, 1980
Stokes	F-Press	Lactose monohydrate, spray-dried lactose, and sodium chloride**OK?	Geoffroy and Cartensen, 1991
Capillary rheometer (with lower load cell and brass push rod)	Carter-Baker Enterprises Ltd./Maywood Instruments Ltd.	Potato starch and granules Sucrose Maltodextrin Sodium chloride	Halliday and Smith, 1997; Ollett *et al*., 1992

Instron Universal Testing Machine	4301	Black pepper	Murthy and Bhattacharya, 1998
Texture Analyzer (Stable Microsystems England)	TA-XT2	Low-fat (2%) milk; instant nonfat milk, instant coffee, ground coffee, cornmeal	Gerhards *et al*., 1998; Yan and Barbosa-Cánovas, 1997, 2000
Instron Universal Testing Machine	4200	Lactose Sucrose Modified cornstarch Butter-oil: single and double encapsulated in lactose, sucrose, and modified cornstarch	Onwulata *et al*., 1995, 1998
High Pressure Machine	Engineered Pressure Systems, Inc.	Instant nonfat milk, spray-dried coffee and freeze-dried coffee	Yan *et al*., 2001

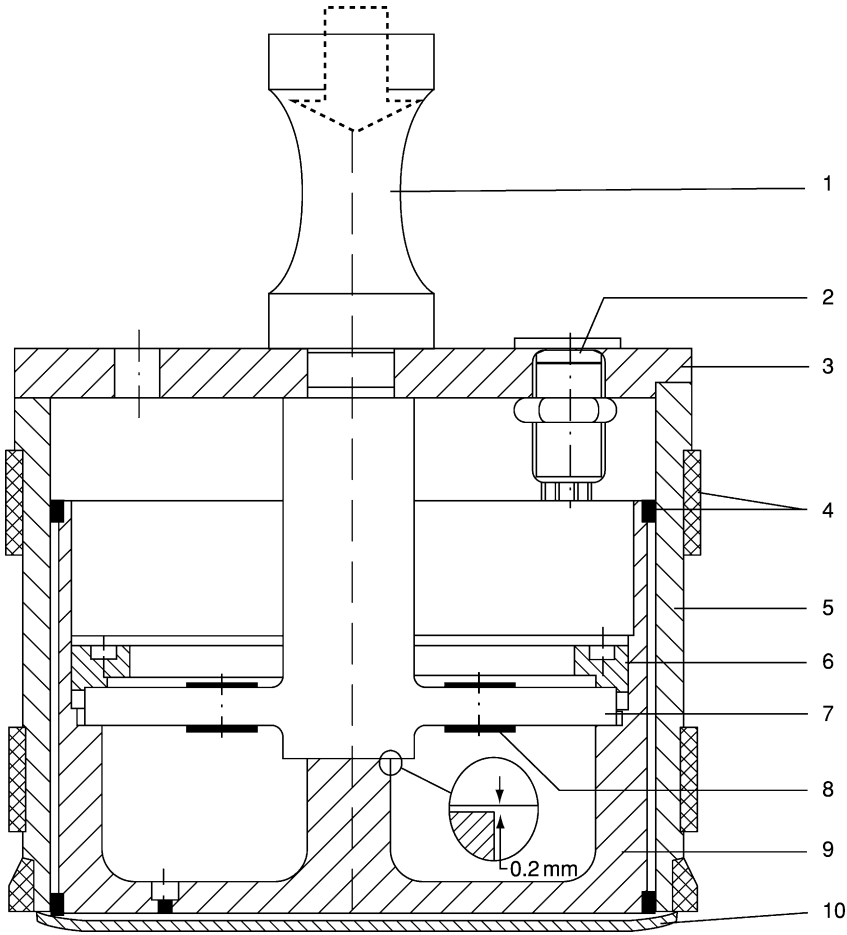

FIG. 7 Device for measuring piston pressure; used with compression cell in place of the standard piston. 1, Pressure knob; 2, miniature receptacle; 3, cover; 4, PTFE rings; 5, guideway; 6, threaded ring; 7, beam; 8, resistance strain gauges; 9, piston; 10, thin foil (drawn to enlarged scale in figure) (from Ehlermann and Schubert, 1987).

4. High hydrostatic pressure method

Isostatic pressing is a technique in which the law of Pascal is applied. In other terms, if a powder is put into an elastomeric mold and sealed and put into a pressure vessel filled with a liquid, the pressure in the vessel will be transmitted to all surfaces in all directions, directly proportionate to the

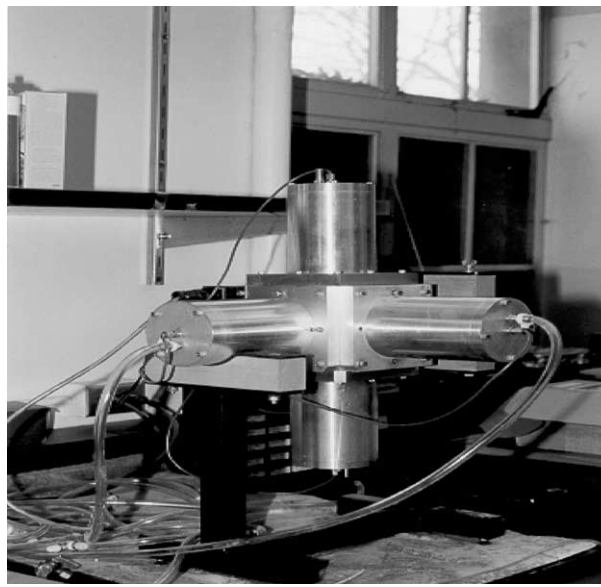

FIG. 8 Triaxial compression tester (from Kamath, 1996).

surface area of the mold (Denby, 1973). Yan *et al.* (2001) introduced the HHP method as a new and useful tester to the traditional compression tester family in food agglomerated powders because of its unique high-pressure and multidirectional forces. HHP acts instantaneously and uniformly throughout the pressure-medium surrounding bag (e.g., water), as shown in Figure 9.

Yan *et al.* (2001), studied how bulk density of instant nonfat milk, spray-dried coffee, and freeze-dried coffee was affected by HHP processing times, particle size, and water activity. The experimental curves for each powder in Figure 10 show that the powder bulk density increased as the pressure increased but remained constant after the pressure reached a critical value of 207 MPa for spray-dried coffee and 276 MPa for freeze-dried coffee at different water activities. The final compressed densities were not significantly different. When the pressure is higher than the critical value, there are no void spaces between the agglomerates or primary particles; even the primary particles are crushed, leaving no open or closed pores within. Bear in mind, it is assumed that the compression mechanisms are the same as those in the confined uniaxial compression tests.

The above final critical densities helped introduce the concept of "ultimate bulk density" (also known as *Barbosa-Cánovas/Yan density*), a value

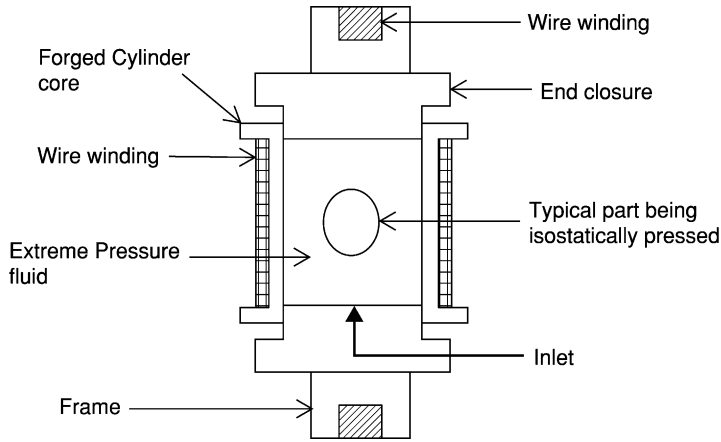

FIG. 9 A high hydrostatic pressure chamber (adapted from Barbosa-Cánovas *et al.*, 1998).

that corresponds to no significant volume change after high-pressure compression. The ultimate bulk density depends on product formulation, physical properties of ingredients, and production conditions. For the same kind of agglomerates, even though they have different initial particle sizes, bulk densities, or water activities, their final compressed bulk densities are not significantly different under the same pressure. Yan *et al.* (2001) also proved that the HHP processing time (at 69 MPa) did not produce any bulk density changes. Therefore, the ultimate bulk density concept could be a promising tool to evaluate powder composition. For example, this value could be used to detect previous composition variations due to changes in product formulation or changes in manufacturing conditions.

5. *Unconfined yield stress test*

The unconfined yield test is a conventional technique that is easily applied to cohesive powders (Buma, 1971; Head, 1982). It is generally used to determine the unconfined yield stress of a specific material. The method is based on preparing consolidated plugs of powder, generally with a cylindrical shape, and then applying an axial load until the powder fails. This load is defined as the unconfined yield stress.

For example, the cohesion of dairy powders has been studied with an unconfined yield test by preparing cylindrical plugs of powder at different particle sizes and moistures. Unconfined yield stress values were obtained as an index of cohesion for whole milk powder and skim milk powder. Dry

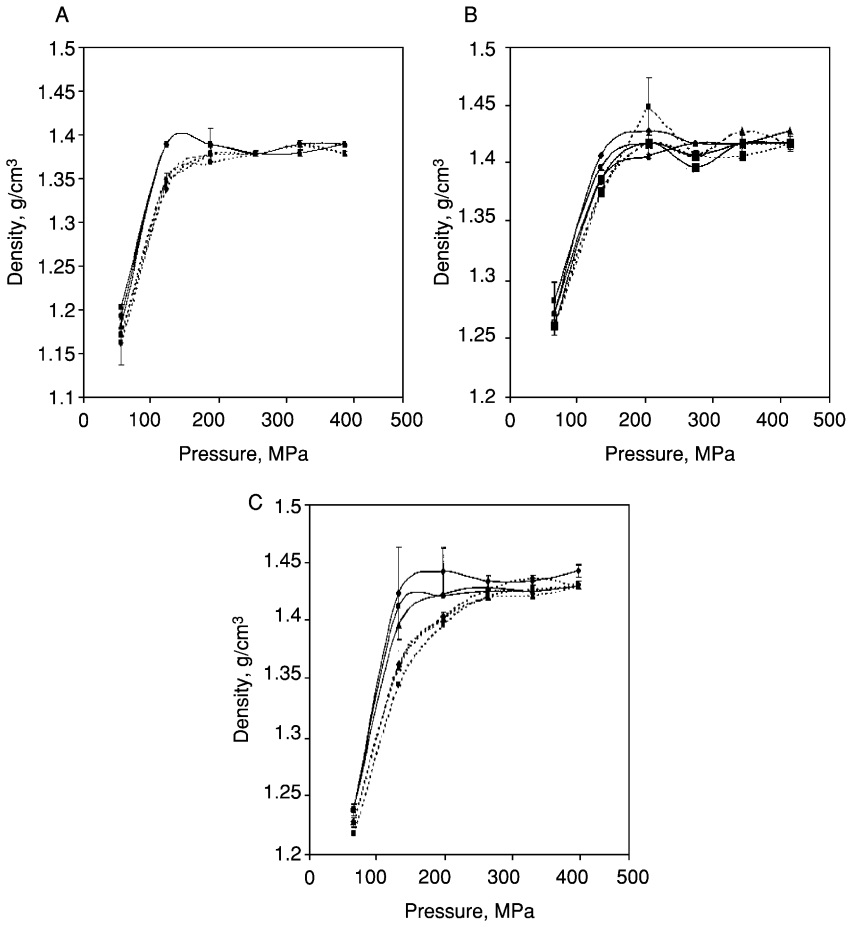

FIG. 10 Pressure-density relation for (A) instant nonfat milk, (B) spray-dried coffee, and (C) freeze-dried coffee; all at different particle-size ranges with two water activities (*dashed line*: $a_w = 0.20$; *solid line*: $a_w = 0.44$) after 5-minute high pressure processing (HPP) treatment at different pressures (from Yan *et al.*, 2001).

whole milk was found to be more cohesive than skim milk with increasing temperature, indicating the influence of fat in the cohesive mechanism for whole milk (Rennie *et al.*, 1999). Milk plugs formed of coarser particle sizes were less cohesive. Furthermore, moisture content produced a lower cohesiveness (<4%; dry basis) and increased cohesiveness (>6%; dry basis). This was explained by a change in lactose from a crystalline state to a rubbery state after reaching the glass transition temperature, which allowed the

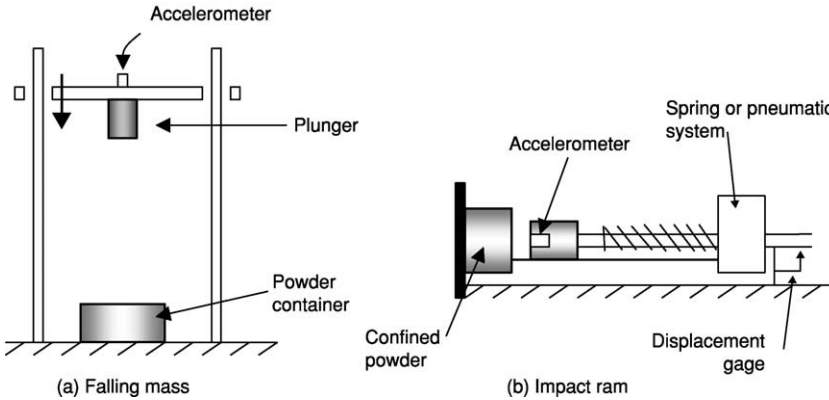

FIG. 11 Two methods used for food powder impact testing (adapted from Mohsenin, 1986 and Hollman, 2001).

formation of liquid bridges, increasing the cohesion of the caked powders (Buma, 1971, Rennie *et al.*, 1999).

Finally, the strength of caked powder has been evaluated with various mechanical methods, ranging from subjective assessment using fingers and drop tests to hardness (Hamano and Sugimoto, 1978) or crushing strength measurements. Rumpf (1961) and Pietsch (1969) have discussed the strength of agglomerates and studied their compression mechanisms (Peleg and Hollenbach, 1984).

6. *Impact methods and crushing tests*

Impact methods test a powder's ability to resist high-rate loading. Different impact methods can be used to characterize powder strength, which include impact of powders with a falling mass, impact tests by ram, and pneumatic dropping of powders on a surface (Hollman, 2001; Mohsenin, 1986). Among these methods, the falling mass and ram methods are compression related (Figure 11). Modern types of impact test equipment (e.g., UTMs) record the load on the specimen as a function of time and/or specimen deflection prior to fracture or particle breakage using electronic sensing instrumentation connected to a computer. Impact resistance is measured in terms of impact energy absorbed by the sample relative to the initial potential energy (gravitational or elastic) of the plunger. As shown in Figure 11, accelerometers are connected to the impact plunger to read the impact forces by use of piezoelectric materials that absorb impact energy, transforming them into an electric signal in response to pressure.

Semiconductor strain gauges added to the plunger can also be used to read a force-displacement profile of the impact event.

Two contact forces are generated in the falling mass test, one between the plunger mass and impacted powder and a second between the powder and container surface at the bottom. In the ram method, the powder is confined and the container walls participate in opposition to the impact response. The falling mass tests depend on the mass selected for the plunger and are designed to determine the maximum drop height that powders will resist before breakage, whereas in the ram test, the pneumatic system controls the impact force ejected. Impact tests can be used to determine food powder coating resistance.

The pneumatic drop impact test was used to study the influence of processing conditions (belt and pneumatic conveying, among other operations) in NaCl crystals (Ghadiri *et al*., 1991). Attrition behavior was studied using a single particle attrition testing rig (to monitor attrition propensity), where particles were thrown vertically in a single downward direction, and a force transducer read the impact force received by the accelerated particle (Hollman, 2001).

Furthermore, these tests can be used for powder attrition testing by measuring the crushing strength necessary to produce a certain number of fines, or conversely, to evaluate the state of breakage (Couroyer *et al*., 2000). In a slow compression test, a close-fitting piston is applied continuously over a fixed period. The percentage of remaining powder greater than a specified particle size is taken as the *crushing strength index* (Bemrose and Bridgwater, 1987). It is not always clear what exactly is measured in uniaxial compression, when compressing agglomerates. One theory behind this test is that by measuring the crushing strength (stress to macroscopic failure), one can estimate the tensile strength of the material. This could be true for highly brittle and isotropic materials in which a compressive stress leads to tensile cracks, which is normal to the maximum principal stress.

7. *Shear cell*

Shear cells can be used to study attrition effects in particles under compression (Ghadiri and Ning, 1997). One example is the direct shear cell, which usually consists of two halves one on top of the other; the one at the top has a replaceable lid that covers the powder and acts like a piston. A schematic diagram of a shear cell is shown in Figure 12. In this case, the compartment at the base is mobile. When a sample is put in the shear cell and compressed under normal force by the lid, the base compartment can be placed in motion by a horizontal shearing force. Once they are filled with the particulate material of known particle size (or size distribution) the test sample is

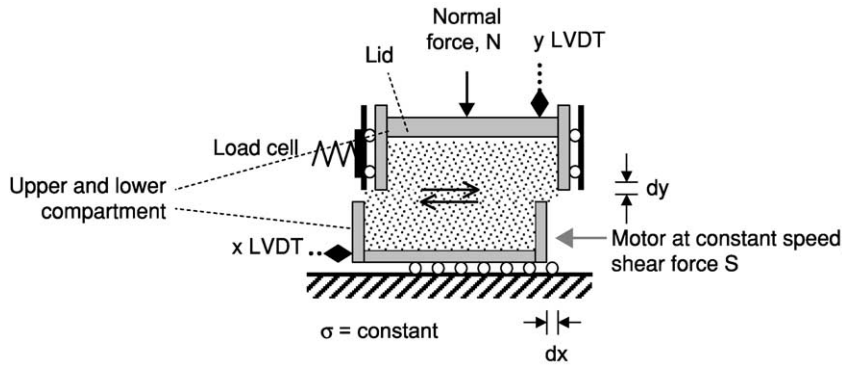

FIG. 12 Shear tester. A constant normal load N is applied over the lid, acting like a piston. A motor moves the lower compartment, and a shear load cell measures the shear force necessary to maintain the upper compartment still. Two linear variable displacement transducers (LVDTs) measure the horizontal and vertical displacements in the cell.

consolidated to become a prescribed packing density. Then, under compression load, a horizontal shear force is applied to the lower cell for a predetermined or standardized period. Particle size distribution is compared before and after the experiment, and fines generated due to interlocking, frictional, and compaction forces are a measure of attrition.

8. Vibration tests

To determine the effect of handling processes, such as jarring, jostling, and vibrating, on the attrition of agglomerates, in a controlled and reproductive manner, particle movement can be introduced by using either a form of resonance or a simple mechanical motion transmitted from a container to the particles within. Bemrose and Bridgwater (1987) described a 40-mm cylindrical drum mounted to a "vibro-saw," which gave a vertical vibration of 6 mm in amplitude at a frequency of 50 Hz. In these tests, the vibration intensity of the particles depended on particle size and density, vibration frequency and amplitude, and depth of the bed. Other parameters for consideration would be particle size distribution and moisture content.

To simulate density changes during handling and transportation, a tap Density Tester (Vankel Industries, Inc., Edison, NJ) that provides vertical vibration is used for vibration tests. A sample with known quantity (weight or volume) is freely poured into a graduated cylinder that rotates and taps simultaneously at controlled speed and amplitude. During the test the sample in the cylinder undergoes volume reduction and attrition as it is exposed

to compaction compression and/or mechanical vibration. After a number of taps (i.e., vertical motions), the level of the sample in the cylinder is recorded and/or the particle size distribution is analyzed. This method is often used to study the compaction characteristics of powders and the attrition tendency of agglomerates. Research has been conducted for instant coffee, milk powders, and other agglomerated food powders (Barletta and Barbosa-Cánovas, 1993a; Barletta *et al.*, 1993a; Malavé-López *et al.*, 1985; Yan and Barbosa-Cánovas, 2001a).

C. INTERPARTICLE ADHESIVE FORCES IN STATIC POWDERS AND AGGLOMERATES

In general, interaction between particles is regulated by the relationship between the strength of the attractive (or repulsive) forces and gravitational forces. Thus, surface attraction forces can have a negligible effect on larger particles (e.g., granular sucrose). Such effects are evident not only in the powder microstructure and appearance of particles, but also in properties like bulk density, compressibility, and flowability, which can be totally altered.

To better understand the compression effects on the microstructure of powder beds in fine and agglomerated powders, one must have a fundamental knowledge of the interparticle forces that intervene (Scoville and Peleg, 1981). For particles in the amorphous rubbery state, it is well known that the following forces cause primary particles to stick together (Hartley *et al.*, 1985; Schubert, 1981): interparticle attraction forces (molecular and electrostatic), interlocking forces, liquid bridges, and solid bridges. During compression, the contribution of adhesive forces can be relatively small. For example, the adhesion force contributions of a silicon model system, used as a reference system for biological friction tests, are 10^{-10} to 10^{-8} N for molecular forces, 10^{-8} to 10^{-6} N for electrostatic forces, 10^{-7} to 10^{-2} for liquid bridges (or capillary forces), and 10^{-5} to 10^{-1} for forces due to excess charges (Scherge and Gorb, 2001). In fact, interparticle forces are inversely related to the particle size (Adhikari *et al.*, 2001; Buma, 1971; Rennie *et al.*, 1999). A diagram showing the strength of agglomerate bonds as a function of particle size is given in Figure 13.

1. Interparticle attraction forces

There are two main types of interparticle attraction forces: Van der Waals (or molecular forces) and electrostatic forces. Van der Waals forces arise from electron motion among dipoles. On a molecular level, they act over very short distances within the material structure. Those forces become particularly significant when the particle is smaller than 1 μm (Hartley

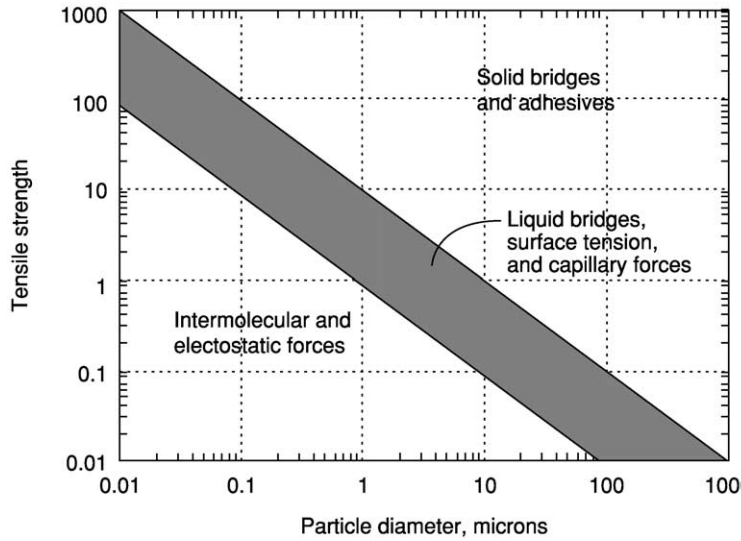

FIG. 13 Strength of bridges needed to hold particles together. The intermolecular and electrostatic forces will not be active above 10 μm (adapted from Rumpf, 1962 and Adhikari *et al*., 2001).

et al., 1985). In fact, smaller particles will have more contact area and therefore more intimate contact in which molecular forces act (Adhikari *et al*., 2001). Electrostatic forces are longer ranging forces that arise from surface changes on particles, provoking charge formation and attraction to different zones. Those forces are present when the material does not dissipate electrostatic charge. Electrostatic forces appear predominantly during mixing and compaction due to triboelectric charging.

2. *Interlocking*

Particles with irregular fibrous shapes or plate-shaped forms can be mechanically interlocked. Mechanical interlocking is used to describe the hooking and twisting of the packed material. By the aid of vibration or pressure, they can reach mutual orientations in which they become physically bound.

3. *Liquid bridges*

Liquid bridges result from the presence of bulk liquid (generally unbound water or melted lipids) between the individual particles. Once a liquid bridge is established, any evaporation of liquid reduces the radii of curvature of

liquid–gas interfaces, thus increasing the forces holding the particles closer together. Liquid bonding can be due to movable liquids (capillary and surface tension properties) and non–free movable binder bridges (viscous binders and adsorption layers). The forces of particle adhesion arise either from surface tension in the liquid–air system (e.g., in a liquid droplet) or from capillary pressure. Capillary pressure is the difference between the pressure of the interior of a liquid strand suspended between two particles and the ambient pressure. This difference is given in the following equation:

$$\Delta p_c = \sigma \times \left(\frac{1}{R_1} + \frac{1}{R_2}\right), \tag{8}$$

where σ is the liquid surface tension in air and R_1 and R_2 are the principal radii of curvature of the liquid bridge, which are functions of the contact angle between the liquid and solid (Schubert, 1981). Therefore, the strength of a liquid bridge depends on (1) factors affecting the contact angles (e.g., composition of solid and nature of liquid solution) and (2) factors influencing the radius of curvature (particle size, shape, interparticle distance, particle roughness, and ratio of liquid to solid in agglomerate).

The composition of the liquid bridge varies in different food materials. The "bridging potential" or "stickiness" is related to factors such as powder moisture, fat or low-molecular-weight sugar content, and shape of particles. For example, viscous liquid bridges may cause flow difficulties in fat-containing powders (Nyström and Karehill, 1996). High water activity acts as a means of providing attractive forces in the form of liquid bridges, due to surface dissolution, liquefaction, or the appearance of a condensed water layer.

Depending on the amount of liquid in the bridges that form a granule, four states of liquid bonding have been identified: pendular, funicular, capillary, and droplet (Lloyd, 1983) (Figure 14). In the pendular state, depending on the powder composition, the strongest forces can be involved;

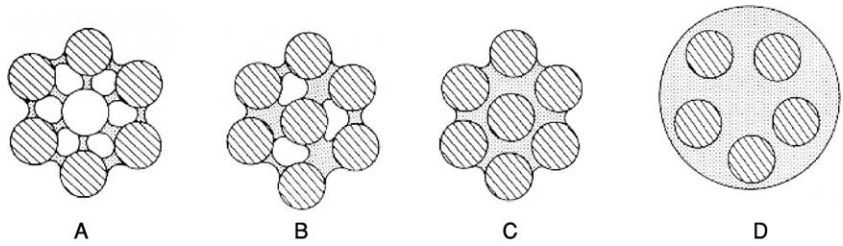

FIG. 14 Types of liquid bonding in granules: (A) pendular; (B) funicular; (C) capillary; (D) droplet (adapted from Lloyd, 1983).

semisolid bridges between the particles are all separate, independent, and centered between the particles at the contact points. If the amount of liquid per interparticle bridge is slightly increased, the bonding force will become less strong and the granule can classify as funicular shaped. At higher liquid contents, all the interstices are filled by capillarity with liquid, and the strength of the granule is due to curvature of the liquid on the surface of each particle. In the last state, the particles are not in contact and the droplet has very little strength.

4. Solid bridges

Solid bridges form as a result of sintering, solid diffusion, condensation, or chemical reaction, of which all are more likely to happen at elevated temperatures after a solution of soluble matter solidifies at room temperature (e.g., sugars or salt). The magnitude of the adhesion force depends on the diameter of the contact area and the strength of the bridge material (Loncin and Merson, 1979). For example, powders containing low-molecular-weight sugar (e.g., in powdered fruits and vegetable powders) can form solid bridges when temperature is decreased and/or moisture is removed by drying of liquid bridges formed in amorphous powders in their rubbery state. Bridging occurs whenever an area of true contact is established between two surfaces, because the interfacial energy is always less than the surface energy.

To determine the prominent interparticle interactions occurring between the particles of a certain food powder under certain moisture conditions, the activation energy of particle bonding can be determined using the Arrhenius plot of tensile strength versus temperature. For example, Lai *et al.* (1986) found activation energy values in egg powders containing corn syrup and NaCl within the range of hydrogen bonding, suggesting that these types of interactions were responsible for cohesion and caking.

D. COMPRESSION AND COMPACTION MECHANISMS

Some properties of materials can vary according to the rate at which stress is applied; some materials are plastic and ductile if the stress is applied slowly but can be elastic or brittle if the stress is applied by impact. The deformation mechanisms occurring during compaction of fines and agglomerated foods depend on the elastic and viscous flow, in addition to ductile yielding and brittle behavior common in pharmaceutical and food compaction processes (Barletta *et al.*, 1993b).

Bika *et al.* (2001) explained how fracture planes can be formed under compressive forces and showed how different stress-strain curves can be for uniaxial tension, uniaxial compression, and isostatic compression

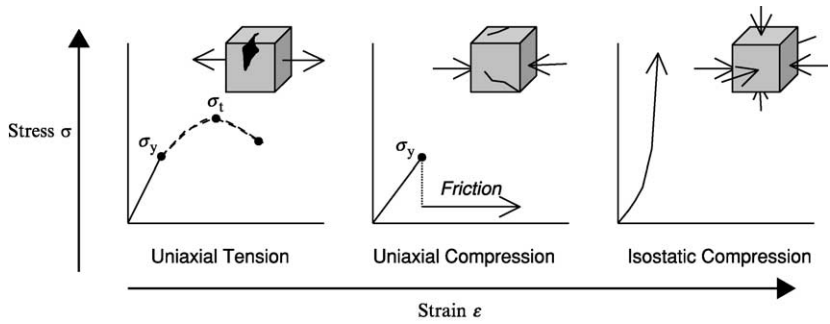

FIG. 15 Characteristic stress-strain behavior of an elastic compressible agglomerate (*solid lines*) in common testing configurations: uniaxial tension, uniaxial compression, and hydrostatic compression (from Bika *et al*., 2001). Yield point σ_y and tensile strength σ_t are indicated.

(Figure 15). Because breakage occurs along cracks in some materials, the breaking point measured by compression is usually higher than when measured by traction; tension enhances the cracks, whereas compression tends to close them up. Compaction mechanisms in brittle agglomerated particles include not only voidage reduction through particle rearrangement and segregation, but also attrition due to impact between particles and container walls.

Because a single agglomerate involves cohesive and adhesive forces, collisions among agglomerates and against the static container walls due to mechanical compression or vibration provide the kinetic energy needed to cause attrition and compaction (Barletta *et al*., 1993a). The strength of an agglomerate can be defined as the strength at which a material either begins to deform plastically or develops macroscopic damage. The compressive mechanisms for fine powders and agglomerates in bulk are different and are described next.

1. Compression mechanisms

a. In fine powders. In the case of fine powders, the compression process takes place in two stages. The first involves particle movement and filling of voids of the same size or a larger size than the particles themselves. The structure of the powder bed can succumb under relatively low pressure up to about 100 MPa, but the particles are not broken or deformed to any significant extent (Peleg, 1978). The packing characteristics of particles or a high interparticulate friction between particles will prevent any further interparticulate movement (Nyström and Karehill, 1996). The second stage involves

filling of smaller voids by particles that have been deformed either elastically (reversible deformation) and/or plastically (irreversible deformation), and eventually broken down (Cartensen and Hou, 1985; Duberg and Nyström, 1986; Kurup and Pilpel, 1978).

The process mainly occurs as a result of friction and interlocking of sliding planes of atoms as a response of the applied stress. The plastic deformation of materials occurs nonhomogenously by means of lattice faults (dislocations) within the crystal structure of materials (Benbow, 1983). Most organic compounds exhibit consolidation properties, undergoing particle fragmentation during the initial loading, followed by elastic and/or plastic deformation at higher loads.

b. In agglomerates. Bulk compression can be broken down into three distinct segments: (1) agglomerate rearrangement to fill the voids of same size or larger agglomerates; (2) agglomerate deformation or breakdown to fill the voids of smaller size agglomerates; and (3) primary particle rearrangement, elastic, and plastic deformation, as well as fracture (Mort *et al*., 1994; Nuebel and Peleg, 1994). The difference between bulk and individual compressions of agglomerates is the bulk's cushioning effect on the particles, thus reducing the amount of fracture. This is common for brittle cellular solid foams. Initially, normal stress varies linearly at very small strains; then linearity of the stress–strain relation ends abruptly followed by an upward concave continuation (Nuebel and Peleg, 1994). It can be assumed that the compression of agglomerates under high multidirectional hydrostatic pressure conditions has the same rearrangement and compaction mechanisms (Yan *et al*., 2001).

Agglomerates with glassy (nonequilibrium) microstructure undergo plastic nonrecoverable deformation before gross failure (Bika *et al*., 2001). Agglomerates flow under the action of stresses during elastic and plastic accommodation through the relative motion of their constituent particles, resulting in bulk deformation or fracture. All mechanical parameters for glassy agglomerates during compression are path and rate specific. Two major reasons explain nonuniformity in the stress transition between agglomerated particles: (1) They are concentrated in preferred paths, constituting a "stress fabric" where some particles experience higher loads and others little or no load, and (2) a distribution of defects (e.g., pores, grains, or internal cracks) may dominate the macroscopic response to stresses (Bika *et al*., 2001).

Both compression mechanisms in fine and agglomerated powders are influenced by particle size and size distribution, particle shape, and surface properties. Potato starch and powdered milk have demonstrated that powders will crackle during compression, that is, change in volume

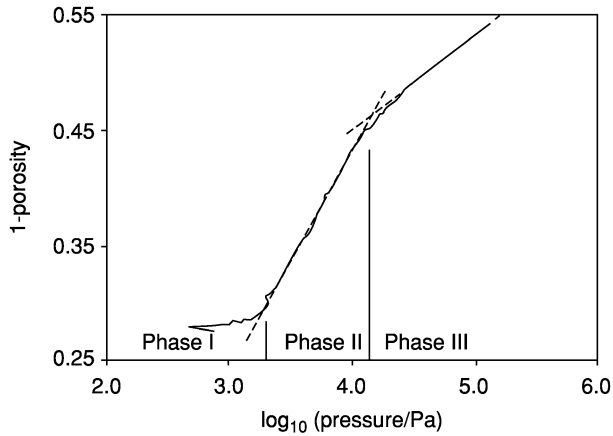

FIG. 16 Typical shape of a compression curve for household wheat flour (type 405) (from Ehlermann and Schubert, 1987).

discontinuously (Gerritsen and Stemerding, 1980). When compressive forces are applied, they are transmitted at the contact points. The magnitude and the direction of the resulting forces at the individual contact points will vary considerably, even if the compression is isostatic. With continuing compression, both the normal and the tangential component of each interparticle force will increase, until at some contact points the material is no longer able to sustain the force and yields. Several materials that crackle during compression have shown considerable compressibility.

The inherent ability of the powder to reduce its volume during compression could affect the amount of interparticulate attraction in the final compact. A decrease in compact porosity with increasing compression load is normally attributed to particle rearrangement elastic deformation, plastic deformation, and particle fragmentation. Scanning electron microscopy (SEM) for the qualitative study of volume-reduction mechanisms has been presented in the literature.

A typical compression curve is shown in Figure 16. During the initial phase the surface of the piston makes contact with the surface of the bulk material. There is practically no change in porosity during this phase and the powder is not yet flowing. During phase II the large voids between particles are eliminated by rearrangement of the particles, breaking the material bridges above. The large pores between particles that cause loose packing are characteristic of cohesive powders. The loose structure of uncompressed powders is due to particle adhesion. Thus, phase II defines the range of pressures of most interest in correlating the results between a compression

test and a shear test. During phase III, moving and rearranging the particles into small regions can achieve further reduction in volume. During this final phase a comminution of particles occurs and a consolidated tablet results. The pressure range of phase III is much greater than that occurring under normal food storage conditions. The evaluation of compression measurements should differentiate between phases II and III (Ehlermann and Schubert, 1987).

2. *Segregation mechanisms*

The segregation process usually occurs in free-flowing compaction systems of particles with a wide particle size distribution. *Particle segregation* refers to the downward migration of smaller particles through a powder bed under motion or vibrating conditions while the coarser particles remain on top. Interparticle bonding impedes segregation (Lindley, 1991; Peleg, 1983), so segregation is less likely to occur in cohesive powders, as fines usually adhere to the surface of coarser particles. Three main segregation mechanisms have been identified (Williams, 1976): trajectory segregation, interparticle percolation or sifting, and rise of coarse particles during vibration or upthrusting. Of these three, the two latter mechanisms can be found in compaction processes.

Interparticle percolation occurs in a bulk in which particles possess a large particle size difference, allowing the smaller particles to drain through the lattice of larger particles due to gravity or motion. Percolation can also take place during particle bed vibration or shear. *Upthrusting* mechanism refers to upward movement of a large particle from the bottom of the bed to the bed surface when the intensity of vibration is suitable or when the large particle, of different composition, is denser than the finer particles. In this case, particle rearrangement causes an increase in pressure in the region below, compacting the material and stopping coarser particles from moving downwards by locking them into position (Williams, 1976).

3. *Attrition mechanisms in food agglomerates*

Some food agglomerated particles such as powdered milk and coffee are brittle and fragile and can easily be broken down during attrition when colliding with each other or against static walls during compaction processes (Barletta *et al.*, 1993a). Attrition can change product appearance, affect agglomerate bulk properties such as flowability and angle internal of friction, reduce a powder's instant properties, and cause segregation with important changes in bulk density (especially during storage) and dust formation related to environmental hazard problems (Barbosa-Cánovas *et al.*, 1987; Barletta *et al.*, 1993b; Malavé-López *et al.*, 1985).

To avoid attrition, processing conditions can be adjusted as a function of the interaction between material properties and processing conditions. Therefore, knowledge of mechanical changes, attrition mechanisms, and morphology is required. Compression tests, in combination with particle size distribution charts, attrition indexes (Barletta *et al*., 1993b), the Hausner ratio (Barbosa-Cánovas and Yan, 2003; Barletta *et al*., 1993b; Hayes, 1987; Malavé-López *et al*., 1985; Yan and Barbosa-Cánovas, 2000), and SEM (Carić, 1994; Kaláb, 1979; Yan and Barbosa-Cánovas, 1997) can provide good background information to characterize this effect.

Three main mechanisms govern the particle attrition process: fragmentation or shattering, surface erosion or abrasion, and a combination of the former denominated chipping (Barletta *et al*., 1993b). Particle shattering indicates the breakage into several midsize particles (relative to parent particles). Erosion characterizes the separation of very fine particles from the surface layer and edges or corners of parent particles that remain slightly smaller. The third attrition mechanism is characterized by partial fracture, which produces small fine particles, plus a ''chipped'' product near the parent size. In this sense, chipping resembles an erosion process rather than a shattering process (Biscans *et al*., 1996). When agglomerated powders are under compressive load, their compaction behaviors will be much different from those of nonagglomerated particulate materials because agglomerates will undergo a relatively higher degree of attrition.

The three attrition mechanisms are in turn governed by different failure modes: brittle, semibrittle, and ductile (Ghadiri, 1997). Brittle failure occurs when internal or surface cracks already exist and is dominant at low elastic deformation at the powder contact surface (Shipway and Hutchings, 1993). Semibrittle failure, at limited plastic deformation, is responsible for flaw initiation and occurs when the impact forces surpass the yield point. In fact, median and radial cracks cause particle fragmentation, and lateral cracks cause chipping. Soft materials are usually ductile and the mechanisms for particulate solids under ductile mode have not yet been elucidated (Ghadiri, 1997).

II. MODELING COMPRESSION AND COMPACTION OF FOOD POWDERS

A. PRESSURE-DENSITY RELATIONSHIPS IN FOOD POWDERS

Many investigators have suggested empirical equations to describe the pressure-density (or pressure-porosity) relationships during compression processes. About 20 equations have been listed for powder compression in powder ceramics (Macleod, 1983) and for other kinds of powders (Peleg,

1983). Some of these include Athy (Chen and Malgham, 1994), Cooper-Eaton (Kurup and Pilpel, 1978; Paronen and Ilkka, 1996), Kawakita (Georget *et al.*, 1994; Paronen and Ilkka, 1996; Ramberger and Burger, 1985), Heckel (Paronen and Ilkka, 1996; Ramberger and Burger, 1985), and Sone (Moreyra and Peleg, 1980). Kawakita, Cooper-Eaton, and Heckel have been widely used for pharmaceutical purposes (Paronen and Ilkka, 1996). Most of these equations were adjusted for particle sizes less than 1 mm (Georget *et al.*, 1994). Some of these models are described below, as well as some applications to food powders. Furthermore, we will show how powder compression mechanisms can be better described by four-parameter equations.

1. *Cooper-Eaton*

A bi-exponential equation can represent compaction of powders, first, by filling the same or larger size voids rather than the initial smaller voids and, second, by filling the smaller voids (due to particle deformation), thus proceeding with elastic or plastic deformation or fragmentation.

$$\frac{(\rho_{bf} - \rho_{b0})\rho_s}{(\rho_s - \rho_{b0})\rho_{bf}} = a_1 \exp\left(-\frac{k_1}{\sigma}\right) + a_2 \exp\left(-\frac{k_2}{\sigma}\right), \tag{9}$$

where ρ_{b0} and ρ_{bf} are the bulk densities at zero stress, ρ_s is the solid density, and at σ, a_1, and a_2 are dimensionless constants, and k_1 and k_2 are constants relative to the stress applied. Dimensionless constants indicate the fraction of the theoretical maximal densification achieved by filling voids of the same size (a_1) and smaller size (a_2) than the actual particles. The model can accurately describe the initial stages of volume reduction providing information about particle surface properties, shape, and size of the densification columns.

2. *Kawakita*

The degree of volume reduction of a powder column as a function of the applied pressure has also been modeled with the Kawakita equation.

$$1 - \frac{\rho_0}{\rho_p} = \frac{ab\sigma}{1 + b\sigma}, \tag{10}$$

where ρ_0 is the initial density, ρ_p is the apparent density at pressure σ, and a and b are constants characteristic of the powder being compressed. Some authors claim that constant a would indicate the maximum volume reduction and describe the compressibility of the powder, and b would describe the volume reduction tendency. However, the physical meaning of

these constants is of questionable significance in many powders (Paronen and Ilkka, 1996).

3. Heckel

Heckel introduced an equation for the densification phenomenon of a powder column following the first-order kinetics relating the powder relative density with compression pressure σ.

$$\ln \frac{\rho_s}{\rho_s - \rho_p} = k\sigma + A, \tag{11}$$

where k and A are constants obtained from a natural log-linear plot. The Heckel equation was used for compaction behavior studies in sodium chloride, sucrose, potato starch, and maltodextrin (Paronen and Ilkka, 1996). It was found that the natural logarithm on the ordinate axis linearized any exponential decreases in porosity and emphasized differences in low porosity. However, the Kawakita equation is valid at low pressure and large intermediate porosity, whereas the Heckel equation gives the best fit at intermediate to high pressure and low porosity.

By evaluating the compression cycle with the Heckel equation for both compression and decompression phases, it is possible to obtain a fairly comprehensive characterization of the mechanisms of volume reduction. For example, the compression cycle was applied to characterize consolidated lactose, sodium chloride, and sodium bicarbonate with 45% moisture. Sodium chloride and sodium bicarbonate were shown to be homogeneous and practically nonelastic, nonporous materials with consolidation behavior similar to metal powders. However, lactose suffered some elastic deformation with some degree of fragmentation and elasticity during the compression–decompression cycle (Duberg and Nyström, 1986).

4. Panelli-Filho

A new model was compared to known compaction equations, including Kawakita and Heckel, for some mineral salts. Panelli and Filho (2001) concluded that Equation 12 best represents the density–pressure relationship for powders, obtaining a linear correlation coefficient close to the unity.

$$\ln\left(\frac{\rho_s}{\rho_s - \rho_p}\right) = A_{10}\sqrt{\sigma} + B_{10}, \tag{12}$$

where A_{10} and B_{10} are characteristic constants. A_{10} represents the ability of the powder to densify by plastic deformation, and B_{10} would represent

the density of the powder at the beginning of the compaction if the rearrangement does not occur.

5. *Sone's compressibility*

Sone's model has been found valid up to a pressure of 4.9 kPa with no expectation of particle yield or breakage, of which the mechanism for powder deformation is described as particle special rearrangement (Barbosa-Cánovas *et al.*, 1987; Peleg, 1978). This model gives a clear sense of the powders' characteristic constants for the description of mechanical behavior (Malavé-López *et al.*, 1985; Moreyra and Peleg, 1980), expressed as follows:

$$Y(\sigma) = \frac{\rho_b - \rho_{b0}}{\rho_{b0}} = C_1 + C_2 \log \sigma, \tag{13}$$

where $Y(\sigma)$ is the density fraction or volume fraction, ρ_b is the bulk density under compression stress σ (unitless and relative to atmospheric pressure), ρ_0 is the powder's bulk density before compression, and C_1 and C_2 are characteristic constants of the powder, C_1 representing the value of $Y(\sigma)$ at a unit stress and C_2 (known as the *compressibility index*) representing the change of relative density with the applied stress.

In some cases, the natural logarithm has been used for the compressibility characterization of food powders. Equation 14 shows the natural semi-log expression for compressibility determination, where density fraction $Y(\sigma)$ is expressed as a function of porosity ϵ between porosity and the solid density ρ_s of particles (Equation 13).

$$1 - \epsilon = a + b \ln(\sigma), \tag{14}$$

where a is a constant and b is the compressibility index (if logarithm is base 10, C_2 is denominated b_{10}). Barbosa-Cánovas *et al.* (1987) used a log–log relationship for the study of compressibility in binary mixtures of food powders:

$$\log \rho_b = A_1 + A_2 \times \log \sigma, \tag{15}$$

where A_1 is the calculated density under unit pressure and A_2 is also defined as compressibility. Molina *et al.* (1990) used this equation for describing compressibility in ground coffee.

Generally, the compressibility of many powders has been correlated with internal cohesion C and to some extent particle deformability. High compressibility is related to low flowability (expressed in terms of cohesion C) under high consolidation stress conditions (Peleg, 1978; Schubert, 1987). Particularly, compressibility has been found to correlate with cohesion C

in many food powders, such as powdered gelatin, powdered onion, and powdered citric acid (Peleg, 1978). Furthermore, increased powder cohesiveness not only increased the compressibility but also decreased bulk density (Moreyra and Peleg, 1980). Results from different confined uniaxial compression tests were analyzed in Equations 13–15 to evaluate compressibility of the following food powders: fine salt, fine sucrose, cornstarch, baby formula, coffee creamer, soup mix, active baker's yeast, instant agglomerated coffee, instant agglomerated low-fat (2%) milk and nonfat milk, instant skim milk, ground coffee, ground corn, cornmeal, lactose, and flour (Konstance *et al*., 1995; Kumar, 1973; Moreyra and Peleg, 1980; Peleg, 1983; Peleg and Manheim, 1973; Yan and Barbosa-Cánovas, 1997, 2000).

Lai *et al*. (1986) studied the compressibility as a measure of flowability of egg lipid and co-dried carbohydrate and salt, as a function of temperature, moisture, and lipid content. Temperature increased compressibility, resulting from the ability of the cohesive powder bed to maintain an open structure supported by the interparticle forces. These softened, plasticized, and extremely weak structures collapsed under very small pressures giving rise to the measured compressibility. Lipid removal neither improved flowability nor yielded reliable results in compressibility.

6. *Three- and four-parameter models*

The existence of such a large number of models is due to the distinctly different mechanisms by which a powder bed deforms. The relative distribution of each mechanism depends on the particle properties (e.g., size, shape, and hardness), magnitude of applied pressure, and stress distribution within the compacted bulk specimen. Therefore, the compressibility distribution pattern in the same powder may be completely different at different load ranges. Three- and four-parameter mathematical models were developed to describe the stress–strain relationship between cellular solids (Nuebel and Peleg, 1994; Swyngedau *et al*., 1991). For example, Swyngedau's (1991) four-parameter equation was successfully applied to describe the sigmoid compressive stress–strain relationships observed in instant agglomerated coffee (Nuebel and Peleg, 1994). This equation is expressed as

$$\sigma = K_1 \cdot \in^{C_1} + K_2 \cdot \in^{C_2}, \tag{16}$$

where σ is the engineering stress (or compression pressure), K_1, K_2, C_1, and C_2 are constants ($C_1 < 1$ and $C_2 > 1$), and $\in$ is the engineering strain (absolute deformation relative to the initial powder height). Nuebel and Peleg (1994) used this model to study the jaggedness of normalized stress versus the strain relationship, using it as a "fingerprint" for instant coffee. Normalized stress was defined as the difference between engineering normal

stress and normal stress obtained from modeling Equation 16 relative to normal engineering stress obtained from the model.

Swyngedau's equation has proven to be a good model descriptor of food powder agglomerate compression (Yan and Barbosa-Cánovas, 1997, 2000). Yan and Barbosa-Cánovas (1997) converted the Swyngedau model into Equation 17, which allows for the direct use of force-deformation readings recorded from a TA-TX2 texture analyzer.

$$F = A_1 \delta^{B1} + A_2 \delta^{B2}, \tag{17}$$

where F is the compressive force, δ the deformation length unit, and A and B are constants. The A units are force over deformation and the B units are dimensionless. The B_1 value from Equation 17, equal to or lower than 1, is a measure of downward concavity of the force-deformation curve at small deformation. B_1 has been related to the first stage of agglomerate compression mechanism, so it measures the degree of particle rearrangement. B_2 (>1) is the measure of the curve's upward concavity at higher deformations and has been related to the last compression mechanism stage in which primary particle rearrangement, deformation, fracture, and densification occur.

This four-parameter model has proven successful in describing the sigmoid-shaped force deformation (Figure 17) over full compression ranges in instant agglomerated coffee, instant agglomerated low-fat (2%) milk, and instant agglomerated nonfat milk after confined uniaxial compression tests (Yan and Barbosa-Cánovas, 1997). Figure 17 describes a typical force-deformation curve obtained in the experiments and different strain zones

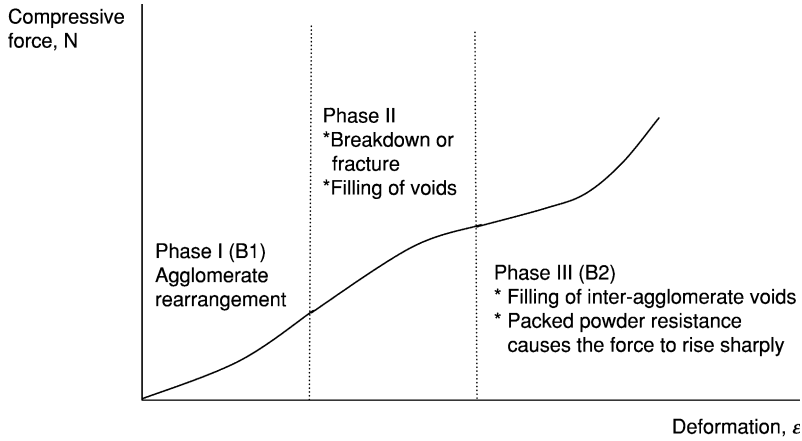

FIG. 17 Force-deformation relationship and description of Equation 16, shown in a four-parameter compression model (adapted from Yan and Barbosa-Cánovas, 1997).

that explain the three steps for the agglomerated compression mechanism described earlier.

B. COMPRESSIBILITY BY CONFINED UNIAXIAL COMPRESSION TESTS

The influence of key parameters, such as particle size and moisture content, in food powder development, manufacturing, and control has been widely studied. Other effects provided using cells with different geometries, mixtures of different size particles, or anticaking agents can also be found in various publications. The following sections summarize a literature review of historical contributions and new research on these effects and the use of compression properties for flowability characterization.

1. Effect of particle size

Particle size is one of the factors greatly influencing the physical properties of particulate systems, such as bulk density, compressibility, and flowability (Barbosa-Cánovas *et al.*, 1987). Equations 13–15 show the effects of particle size on the compression behavior. Yan and Barbosa-Cánovas (1997, 2000) found that particle size played an important role in affecting compressibility (C_2) changes in three agglomerated food powders (low-fat [2%] milk, instant nonfat milk, and instant coffee) and two noninstant powders (ground coffee and cornmeal). For the three agglomerated powders, the compressibility (C_2) increased with particle size, given that larger agglomerated particles have a lower bulk density because of larger voids between and within agglomerates. Meanwhile, Rennie *et al.* (1999) found that the particle size of dairy powders had a markedly decreasing effect on cohesion. In the case of cohesive agglomerated dairy powders, it can be deduced that less cohesive materials with larger particle sizes are more easily compressed (i.e., will show increased compressibility).

Molina *et al.* (1990) obtained a good fit in Equation 15 and used compressibility (A_2) to evaluate the particle size influence in ground coffees for two brands. Compressibility values were considerably different for two brands at a given size. Factors such as chemical composition, roasting, and grinding conditions may affect compressibility values as well. However for each brand tested, compressibility did not vary for coarser fractions (500–2360 μm). The truly fine fraction (180–250 μm) formed a low-density open bed structure, which offered a higher compressibility. Therefore, a combination of high compressibility and low density in fine powders proved to be a good indicator of fine powder cohesiveness.

Yan and Barbosa-Cánovas (2000) found that ground coffee and cornmeal fit Equation 13 and powder compressibility (C_2) was affected by particle size. Finer particles gave higher compressibility, and larger particles had higher loose bulk density, coinciding with the research of Molina *et al.* (1990). Larger particles mainly underwent the two steps in the fine powder compression process explained earlier. However, for smaller particles, the compression process stopped at the first compression step filling the space voids because of the bulk high porosity.

Particle size was also evaluated in encapsulated materials by comparing single- and double-encapsulated butter oil powders in sucrose (<500 μm and 1000 μm, respectively) by Onwulata *et al.* (1998). Compressibility as a measure of cohesion and mechanical strength at various loads increased for both single-encapsulated powders and double-encapsulated powders. However, larger powders were more impeded in flow than single-encapsulated ones.

2. *Effect of moisture*

Moisture content is the key analysis made for development, production, and quality control of food powder products. Moisture sorption and diffusion not only affect the shelf life of a product but also cause physicochemical changes such as sticking, collapse, caking, agglomeration, loss of volatiles, browning, and oxidation. These changes are of real concern for the food powders industry because of the economical losses they can bring during the production and storage of these products. Amorphous and semicrystalline powdered materials are capable of absorbing and desorbing water. Mechanical properties related to the interaction of food powders can change according to shifts in the glass transition temperature of a product because of its water content variation. For example, although sodium chloride and sucrose are crystalline materials, they can exhibit ductile and brittle deformation during compaction processes depending on their water content or the relative humidity in the system. On the other hand, other products can present ductile behavior even when dry. Potato starch is semicrystalline and the maltodextrin is rubbery in dry conditions or at low moisture contents (Ollett *et al.*, 1992).

Moreyra and Peleg (1981) studied the influence of water activity on the compressibility of baby formula, ground bran, powdered onion, powdered sucrose, and granular sucrose. All powders showed increasing compressibility (C_2) with increasing water activity and a corresponding decrease in bulk density. Bulk density decreased, because these are cohesive materials with strong interactions such as solid and liquid bridges, enabling the formation of an open structure due to water absorption (Peleg *et al.*, 1973). Barbosa-Cánovas *et al.* (1987) worked with binary mixtures of granular malic acid,

citric acid, granular sucrose, precipitated isolated soy protein, and domestic cornstarch. Compressibility values (C_2) were raised with increasing water activity in fine or coarse nonagglomerated powders and even in binary mixtures.

After proving that whole egg powder with corn syrup and salt was more hygroscopic than whole egg powder alone (higher fat content and protein hydrophobicity), Lai *et al.* (1986) studied the effect of moisture content on compressibility (C_2). Moisture content (3–14% dry basis) had almost no effect on the compressibility of whole egg powder, because a higher concentration of lipids was the dominant factor influencing flow properties. However, whole egg powder mixed with corn syrup and salt had a maximum compressibility of about 4.0% dry basis moisture. At this maximum, an increase in bulk density and a decrease in tensile strength were observed.

Ollett *et al.* (1992) found that amorphous components such as maltodextrin retained a relatively high water content, compared to crystalline materials like sodium chloride, which showed less sensitivity to water content. The Heckel equation (Equation 11) was used to study the effect of increased water, which showed a decrease in deformation stress. The most extensive effects due to water content were exhibited by potato starch, which spanned a large range of stresses. The elastic response of potato starch was particularly sensitive to water content when the material approached glass transition (moisture content 20% wet basis). At low water contents (5% wet basis), the starch granules were glassy and predominantly ductile and only small elastic deformations occurred. At higher water contents (~25% wet basis), the starch granules were rubbery and allowed for extensive elastic deformation resulting in relative densities closer to 1. The lack of crystallinity and lower molecular weight of maltodextrin caused it to form viscous liquid rather than a rubbery solid at water contents higher than 10% (wet basis).

Ollett *et al.* (1992) also observed that particle failure and subsequent rearrangement was involved during compaction of sucrose and sodium chloride. The effects of water content were greatest for potato starch and sucrose, of intermediate value for the maltodextrin and least for sodium chloride. Deformation stresses determined from the Heckel analysis of compaction data decreased with increasing water content. This was interpreted in terms of plasticization for the amorphous materials, whereas for crystalline materials, lubrication effects in the rearrangements following particle failure were invoked.

Gerhards (1998) characterized instant coffee agglomerates at different equilibrium relative humidities. Dry agglomerates ($a_w = 0.11$) had a highly jagged relationship typical of brittle materials, while moist agglomerates ($a_w = 0.69$) had a smooth, concave upward curve characteristic of a

plasticized solid. Stiffness values (slope of plot of local peak force during compression vs distance) remained practically constant in a water activity range of 0.11–0.57 and dropped sharply from 0.57 to 0.69. Particles dissolved and their stiffness assessment became irrelevant because of particle plasticization.

Halliday and Smith (1997) also studied the compaction properties of potato starch and potato granules using the Heckel plot with pressure applied between 1 and 85 MPa, and the Heckel equation (Equation 11) fit to the linear portion of the data. The deformation stress obtained from the equation showed a decreasing tendency in the water content increments. Because starch has a glass transition temperature close to ambient temperature, when the water content is about 20% (wet basis), a particularly sensitive elastic response was observed in the polymer when increasing water activity. The deformation mechanisms occurring during compaction were associated with elastic and viscous flow, in addition to ductile yielding and brittle behavior. At low water contents of about 5% (wet basis), the starch granules are glassy, only small elastic deformations occur, and the behavior is predominantly ductile. At high water content (~25% wet basis), the starch granules are rubbery and allow extensive elastic deformation.

The effect of water activity in coarse non-cohesive materials was studied by Murthy and Bhattacharya (1998) in black pepper seeds (particle size ~5 mm) with a nearly spherical shape (roundness values close to 1). The bulk density of the seeds increased marginally with the increase of moisture content, filling the pores and surface cracks without an increase in volume. At low compression force, the seed offered little resistance to compression, and exhibited a linear elastic behavior. The soft outer coat offered little resistance during initial stages of compression. Once the outer coat was compressed, the inside hard core offered considerable resistance, showing plastic behavior until reaching the failure point when the seed suddenly ruptured into two segments. Moisture altered the mechanical strength of the product by plasticizing and softening the starch protein matrix. As the moisture content was increased (11–15% dry basis), the failure force decreased markedly with an accompanying decrease in the deformation modulus. A high moisture level (31% dry basis) showed a much higher deformation extent, increasing the linear strain limit and decreasing the deformation modulus.

Yan and Barbosa-Cánovas (2000) worked with nonfat milk, low-fat (2%) milk, and instant agglomerated coffee at three water activities, and they proved that the compressibility (C_2) tends to decrease slightly as water activity increases. However, this variation in water content is not as significant as the effect of particle size (Figure 18). At lower water activities, compressibility was higher because of the agglomerated particles' higher

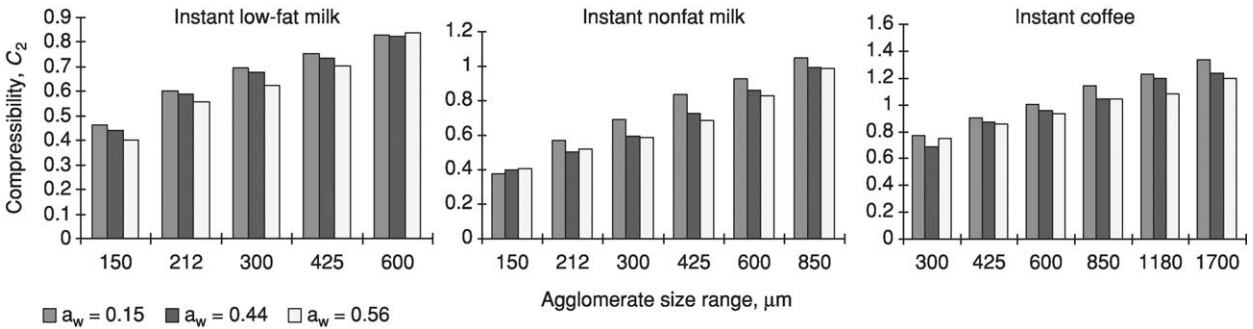

FIG. 18 Powder compressibility C_2 for (A) instant low-fat milk, (B) nonfat milk, and (C) instant coffee, at three a_w values and different size ranges (gray: $a_w = 0.15$; black: $a_w = 0.44$; white: $a_w = 0.56$) (adapted from Yan and Barbosa-Cánovas, 2000).

brittleness. For instant low-fat milk, the compressibility at different water activity levels is significantly different. For both instant nonfat milk and instant coffee, there is no significant difference in compressibility between $a_w = 0.44$ and $a_w = 0.56$, but there is a tendency toward decreased compressibility with increased moisture content.

3. *Effect of cell geometry*

If different researchers used cells of various dimensions for compressibility evaluation, results might not be comparable. To prove this, Yan and Barbosa-Cánovas (2000) investigated how compressibility (C_2) values statistically responded at compression cells of different geometry. Cylindrical confined uniaxial compression cells of diameters 10, 21, and 30 mm and depths 20, 40, and 60 mm, respectively, were used for the experiments. Results showed that the cell diameter and compression bed height had significantly different effects on the compressibility of ground coffee and cornmeal powders.

In particular, compressibility of both powders at bed heights of 20 mm was significantly different from compressibility observed at bed heights of 40 or 60 mm. Furthermore, the lowest diameter cell of 10 mm yielded significantly different compressibility values from cells with larger diameters. It was also observed that the larger the bed height, the more variable the compressive density was along the vertical cylindrical axis. Hence, compressibility evaluation should only be carried out in compression cells having the same dimensions. Standards for powder compressibility studies should include specifications for cell geometry.

4. *Effect of particle size mixtures*

If the bulk densities of coarse and fine components are ρ_c and ρ_f, respectively, then the maximum possible density ρ_{max} of the mixture, provided each component retains its bulk characteristics, is given by (Barbosa-Cánovas *et al.*, 1987)

$$\rho_{max} = \rho_c + \xi \cdot \rho_f, \tag{18}$$

where ξ is the voids ratio. Barbosa-Cánovas *et al.* (1987) evaluated the compressibility of binary combinations of granular malic acid, granular sucrose, precipitated isolated soy protein, and domestic cornstarch mixed at proportions 1:3, 1:1, and 3:1. Model Equations 13 and 15 were used to describe compressibility. Results indicated that the compressibility of mixtures made of powders having similar solid and bulk densities remained unchanged in any combination. When density of components differed

greatly, the mixture's compressibility was higher than that of its components because of the increased particle cohesion and/or plasticization effects (lower density because of an open structure formation). However, no correlation could be found between the effects of the mixture composition on compressibility. In particular, the granular sucrose–cornstarch mixture was the only one among all the studied mixtures showing higher bulk density values than the pure ingredients. This was probably a result of packing, where larger particle (granular sucrose) voids were filled with smaller particles (cornstarch) or there was adhesion of fine particles to carriers (assuming an ordered mixture).

Yan and Barbosa-Cánovas (2000) also studied the effect of mixture conditions on particles, using ground coffee and cornmeal within different size ranges at proportions 1:3, 1:1, and 3:1 on their compressibility (C_2) values. Compressibility results for each mixture showed a linear relationship with that of the monosize particles. The relationship can be described as

$$C_{\mathrm{MIX}} = W_{\mathrm{A}} \times C_{2\mathrm{A}} + (1 - W_{\mathrm{A}}) \times C_{2\mathrm{B}}, \tag{19}$$

where C_{MIX} is the compressibility of the mixture, W_{A} is the percentage of powder with a size range A by weight, and $C_{2\mathrm{A}}$ and $C_{2\mathrm{B}}$ are the compressibility of powder with size ranges A and B, respectively. This equation can be useful for approximating the compressibility of a mixture of two particle size ranged materials and for calculating the material needed to obtain a mixture with an expected compressibility and/or flowability.

5. *Compressibility for anticaking agent effect evaluation*

Flow problems are mainly dependent on interparticle/intraparticle forces, powder particle size and shape, and moisture and fat content. Conditioners (or anticaking agents) enhance powder flow by reducing interparticle force cohesiveness and compressibility while increasing bulk density (Peleg and Manheim, 1973). Peleg *et al.* (1973) showed that as concentrations of stearate or silicate (added to sucrose) were increased from 1% to 3%, there was no reduction in cohesiveness at agent concentrations of 1–2%, but cohesiveness decreased as more flow conditioner was added.

Hollenbach *et al.* (1982) measured the effectiveness of four anticaking agents for diminishing interparticle interactions in fine-powdered sugar, a fairly cohesive material, using compressibility obtained from bulk density values of the mixtures. The addition of GRAS anticaking agents such as silicon oxide, sodium aluminum silicate, tri-calcium phosphate, and calcium stearate (0.1–2%) provided an increase in loose bulk density, depending on the agent and concentration. The presence of the conditioner physically separates the particles, reducing interparticle forces and interfering with

the liquid bridge formation, and forming a denser structure by randomly filling some voids in the powder bed, depending on the geometrical characteristics of the particles. Because this structure was denser, attaining noncohesive powder characteristics, a decrease in compressibility was observed wherein there was no bed structure formation because of interparticle forces.

Compressibility values have also been suggested as an index for anticaking agent selection in a certain food powder system. In this case, selection studies must be within temperature and moisture working ranges. Particle shape and area can give additional information about stickiness behavior for anticaking selection.

Molina *et al.* (1990) studied the effect of adding Hubersorb-600 (0.5%) on the compressibility (A_2) of ground coffee. Unlike in crystalline powders (e.g., ground sucrose or salt), the admixture of the conditioner at 0.5% concentration did not drastically alter the coffee's density and compressibility. However, a new research opportunity was opened for the addition of selected conditioners in coffee packed in bags or stored in bulk to protect its flavor and its physical stability. Konstance *et al.* (1995) studied the addition of 2% Sylox anticaking agent to milk fat powders encapsulated in sucrose, lactose, and all-purpose flour, finding the agent effective in reducing compressibility (C_2).

Onwulata *et al.* (1995) studied the effect of flow conditioners calcium stearate, aluminum silicate, and silica added at different concentrations on bulk density, flow and mechanical properties of lactose, sucrose and modified cornstarch, as well as milk fat encapsulated in the same materials. Each flow conditioner was effective in reducing compressibility (C_2) of the powders when applied at 1% concentration. Compressibility of the remaining unencapsulated powders continued to decrease with added flow conditioner. However, in the case of encapsulated butter-oil powder, the only effective additive was silica (2%), which resulted in a 35–70% decrease in compressibility. The most notable effect was observed with butter-oil powder encapsulated in lactose where reductions in compressibility resulted and an increase in powders flowability. The stearate resulted in flow retardation of all powders studied.

6. *Characterizing flowability*

Peleg (1978) mentions that the characteristic compressibility can be used as a parameter to indicate flowability changes, because compressibility (Equations 13–15) has been related to cohesion C. The more compressible a material is, the less flowable it will be (Carr, 1965). This relationship has been found from experiments on limestone, powdered sugar, semolina, and flours at different particle sizes, size distributions, and moisture contents.

TABLE II
FLOW FUNCTION QUANTITY (f_{fc}) (SCHUBERT, 1987) AND COMPRESSIBILITY (C_2) USING THE MODIFIED UNIAXIAL COMPRESSION TEST METHOD (EHLERMANN AND SCHUBERT, 1987) CAN BE USED TO CHARACTERIZE FLOWABILITY

Flowability	Flow function, f_{fc}	Compressibility, C_2
Nonflowing	<2	>0.02
Cohesive	<4	>0.06
Easy flowing	<10	>0.10
Free flowing	>10	

Hollenbach *et al.* (1982) showed compressibility to be a more reproducible index than parameters such as angle of internal friction or unconfined yield stress obtained from the Jenike shear test (ASTM D 6128).

Conversely, Ehlermann and Schubert (1987) sustained that compressibility results from materials of different composition cannot be compared and that flowability characterization through compressibility must be made specifically for each food variety. Moreover, confined uniaxial compression is a "simple" compression test that provides an approximate measure of the flowability of powders. Therefore, it is not suitable for silo design but may prove to be a convenient method for process control in any food laboratory (e.g., to evaluate particle cohesion). Table II offers a range value definition for flowability classification by comparing flow function (ratio between the maximum consolidation stress and unconfined yield stress) with compressibility.

Yan and Barbosa-Cánovas (2000) used the criteria defined earlier to characterize powder flowability with compressibility measurements in ground coffee and cornmeal. Coarser ground coffee or cornmeal had a lower compressibility and thus better flowability than the finer grinds. However, the same conclusion cannot be drawn in porous agglomerates because compression mechanisms are different. Compressibility in agglomerated powders such as instant coffee and milk increases with particle size because of increased brittleness. In this case, free-flowing characteristics of agglomerates are attributed to a decrease in stickiness. Thus, the cohesion C is not applicable to flowability in agglomerated brittle powders.

C. COMPACTION IN FOOD POWDERS

The purpose of food powder compaction tests is primarily to simulate density changes during handling and transportation. Subjecting a powder to vibration or impact usually results in its compaction. During vibration of

a powder column, the density of the compact usually approaches an asymptotic value determined by the vibration amplitude and frequency. This behavior has been explained with indexes such as the Hausner ratio index and other compaction models. Models developed for the study of attrition and segregation are presented in the following sections.

1. *Hausner ratio*

The Hausner ratio (Grey and Bedow, 1969) has been introduced as an index to measure the amount of bulk density change caused by compaction and has been used to indicate the presence of attractive forces and friction. Malavé-López *et al.* (1985) defined the Hausner ratio as the relation between asymptotic and initial bulk density (Equation 7):

$$H_R = \frac{\rho_\infty}{\rho_0}, \tag{20}$$

where H_R is the Hausner ratio, ρ_∞ is the asymptotic constant density after a certain amount of taps, and ρ_0 is the initial bulk density of the sample. A more direct expression widely used to evaluate flow properties calculates powder volume changes in a graduated cylinder after a certain period of time or number of taps (Hayes, 1987):

$$H_R = \frac{\rho_n}{\rho_0} = \frac{V_0}{V_n}, \tag{21}$$

where n is the number of taps applied to the sample, ρ_n and ρ_0 are the tapped and loose bulk density, and V_0 and V_n are the loose and tapped volume, respectively. H_R can be used to evaluate the flowability from a classification constructed by Hayes (1987), similar to one made by Jenike on the flow function. If the Hausner ratio is 1.0–1.1, the powder is classified as free flowing; 1.1–1.25, medium flowing; 1.25–1.4, difficult flowing; and >1.4, very difficult flowing. The advantage of this method is the simplicity of the instrumentation (Sone, 1972) and the test performance. However, the Hausner ratio may not be a reliable flowability index for most food powders (Peleg, 1978). Experimental results may depend on the test procedure (e.g., number of taps) and the particle size of the tested bulk because compaction patterns of food powders, including the asymptotic density, depend on the vibrations/impacts regimen, which might change the Hausner ratio or vary it dramatically.

Chang *et al.* (1998) evaluated cohesion, angle of repose, and the Hausner ratio of different mixtures of a starchy powder (potato starch) and a proteinaceous powder (wheat protein) at different water activities. As the water

activity of each mixture increased, the tapped bulk density, Hausner ratio, angle of repose, and cohesion increased. However, loose bulk densities decreased at different water activities. Mixtures with higher protein content had a notable tendency to aggregate, indicating increased cohesion. The authors developed empirical equations, which included the tested bulk properties for relating fundamental properties of a powder with the relative humidity of the environment.

2. *Compaction models*

Sone (1972) proposed the following relation between bulk density changes and number of taps to model compaction by tapping (Barletta *et al.*, 1993b):

$$\rho_\infty - \rho_n = C \exp(-\frac{n}{K}), \tag{22}$$

where ρ_∞ is the asymptotic bulk density, ρ_n is the density after n taps, and C and K are constants. Another model relates the volume (or density) reduction fraction $Y(n)$, similar to the one defined in Equation 4 as a function of the number of taps n (Peleg, 1983):

$$Y(n) = \frac{V_0 - V_n}{V_n} = \frac{\rho_{bn} - \rho_{b0}}{\rho_{b0}} = H_R - 1 = \frac{n}{A + Bn}, \tag{23}$$

where ρ_{b0} and ρ_{bn} are the respective densities and A and B are constants. The Hausner ratio may be adjusted to parameter B in Equation 23. The Hausner ratio, defined in Equation 20, can be related to a particular B representing the asymptotic value of $V(n)$ [i.e., when $n \to \infty$, $V(n) \to 1/B$].

$$H = \frac{\rho_\infty}{\rho_0} = \frac{B - 1}{B} \tag{24}$$

A single exponential model proposed by Malavé-López *et al.* (1985) is expressed as

$$Y(n) = C[1 - \exp(-\frac{n}{N})], \tag{25}$$

where C is a constant, n is the number of taps, and N is a constant characteristic of the system. Equation 26 is a double-exponential model proposed by Barletta *et al.* (1993b) to describe coffee agglomerate compaction during tapping:

$$Y(n) = C_1[1 - \exp(-\frac{n}{N_1})] + C_2[1 - \exp(-\frac{n}{N_2})], \tag{26}$$

where *C*s and *N*s are constants. Furthermore, a four-parameter model (Barletta *et al*., 1993b) where *A, B, C*, and *D* are constants is expressed as

$$Y(n) = \frac{n}{A + Bn} + \frac{n}{C + Dn}. \tag{27}$$

The compaction characteristics of agglomerated coffee during tapping were studied by Barletta *et al*. (1993b) using some of the models mentioned earlier. They found that the three- and four-parameter models have shown considerable improvement in fitting experimental data, compared to Equations 22 and 23. To describe the voidage reduction phenomenon of agglomerated powders, three- and four-parameter models are more suitable because they include compaction and attrition effects.

3. *Comparison of mechanical compression and vibration by tapping*

Malavé-López *et al*. (1985) studied the compaction characteristics by observing the vibration and mechanical compression of different kinds of powders, such as instant coffee, wheat flour, rye flour, cornstarch, and soy protein powders, all with different cohesion properties. No clear relation was found between the Hausner ratio and the mechanical compressibility, at least under the test conditions reported in this work. However, H_R was found to be useful in qualifying the maximum powder compressibility under vibration (Malavé-López *et al*., 1985).

Mechanical compressibility in compression tests is, at least partly, a result of the collapse of an initial open bed structure (supported by cohesive interparticle forces or interparticle liquid bridges), which is totally and irreversibly destroyed. In the case of vibrational tapping tests, the "openness" of the bed structure can be recovered at least to some extent. Thus, different mechanisms yield different tendencies in the compressibility measured by tapping and mechanical compressibility.

4. *Compaction and segregation*

Barbosa-Cánovas *et al*. (1985) studied segregation tendencies in some food mixtures during vertical and horizontal vibrations and showed that segregation occurs not only in free-flowing powder mixtures, but also in some cohesive powder mixtures. Segregation intensity depends not only on the mixture composition, particle size, and mechanical vibration history, but also on whether the mixture is "ordered." Barbosa-Cánovas *et al*. (1985) used the segregation index S_{index} after the mixture was subjected to vibration or tapping.

$$S_{index} = \sqrt{\frac{\sum_{i=1}^{n} W_i (X_i - X)^2}{\sum_{i=1}^{n} W_i}}, \tag{28}$$

where W_i is the weight of powder in the ith ring of a vibrating multisplit cell, X_i is the concentration of a given component, and X is the mean concentration of the component in the mixture. Segregation tendency in a starch–sugar mixture was inhibited by attractive interparticle forces at certain mixture ratios, whereas a sugar and instant coffee mixture did not show a cohesion effect, segregating almost completely. It was also noticed that increasing the vibration frequency intensified the mixture's segregation.

Percolation velocity during compaction is affected by the particle size ratio and shape (Shinohara, 1997). Bridgwater *et al.* (1969) developed a mathematical model for this kind of radial dispersion, which was proposed by Shinohara (1997).

$$\ln \frac{N_0}{N_0 - N} = \frac{R}{4E_R t}, \tag{29}$$

where N_0 is the total number of percolated small particles through a packed bed and N is the number of small particles with centers inside the radius R at compaction time t.

Segregation during angle of repose measurement in the slope being formed is especially relevant, given the importance of this test in flow characterization. Percolation is the governing mechanism during the formation of a heap, where the fine particles tend to flow easily to the center. When uniformly sized particles of different materials are mixed, each with different angles of repose, the material with the steepest angle will tend to concentrate in the center, while the one with the flatter angle will concentrate in the outside surface of the heap.

5. *Attrition kinetics*

Particle size analysis is useful for assessing attrition because both fragmentation and fine formation yield separate particle populations with different sizes. Production of midsize particles by means of shattering will lower the particle population's mean size and increase its size spread, as formation of fines through surface erosion will make the overall size distribution bimodal or multimodal. Barletta *et al.* (1993b) summarizes the different size distribution patterns in attrition resulting from the predominant attrition mechanisms and reviews the different models that fit these distributions.

In studying the attrition kinetics of powder compaction through tapping, one of the most convenient ways to organize attrition data is to plot the

percentage of fines retaining original size as a function of characteristic time or number of taps (Barletta *et al.*, 1993a; Malavé-López *et al.*, 1985). Several kinetic models have been proposed to characterize these curves. For example, a good two-term exponential model was found that describes the attrition kinetics of agglomerated coffee with original particle size larger than 16 mesh or 1180 μm (Malavé-López *et al.*, 1985).

$$W_{R/o} = F\ \exp(-A_1 n) + (1 - F)\ \exp(-A_2 n), \tag{30}$$

where $W_{R/O}$ is the weight ratio between agglomerates retaining original particle size and the total sample, n is the number of taps, and F and $(1-F)$ are the fractions of material that have undergone attrition at rates A_1 and A_2, respectively. A nonexponential model was also suggested:

$$W_R = 1 - \frac{n}{B_1 + B_2 n}, \tag{31}$$

where W_R is the weight fraction of particles retaining original size after n taps, and B_1 and B_2 are constants. If $n \to 0$, $1/B_1$ can be considered as the initial attrition rate, and $n \to \infty$, $1/B_2$ as the asymptotic weight fraction of material under attrition. This nonexponential model has some advantages over exponential models. First, it has a residual (asymptotic) fraction of agglomerates that survived the attrition test, whereas exponential models imply that all agglomerates of original size will disappear. Second, it generally fits the experimental data quite well, and its two constants can be easily calculated in regression models (Barletta *et al.*, 1993a).

The attrition index A_I (Equation 32) proposed by Barletta and Barbosa-Cánovas (1993a) was found suitable for studying agglomeration and the effects of agglomerate size and water activity on the attrition kinetics of agglomerated coffee and nonfat milk (Yan and Barbosa-Cánovas, 2001a).

$$A_\mathrm{I} = Cn^{\alpha}, \tag{32}$$

where A_I is the attrition index, C and α are constants, and n is the number of taps. A_I is defined as

$$A_\mathrm{I} = \frac{F}{R}, \tag{33}$$

where F is the weight fraction of fines generated in the attrition test and R is the size stability related to agglomerates retaining original particle size. R can be calculated by the following formula:

$$R = \frac{\sum_{i=1}^{i=L} W_i S_i|_{T=n}}{\sum_{i=1}^{i=L} W_i S_i|_{T=0}}, \tag{34}$$

where W_i is the amount of material retained on each sieve, S_i is the normalized sieve opening size, and T is the number of taps.

Yan and Barbosa-Cánovas (2001a) compared the Hausner ratio to the attrition index A_I under different test conditions in agglomerated coffee and milk. The Hausner ratio was closely related to the attrition index at high numbers of taps (>5000 taps) due to agglomerate attrition. However, at low numbers of taps, the Hausner ratio only represented a volume reduction due to agglomerate rearrangement in the test cylinder. Therefore, it was suggested that the Hausner ratio could be used as a simple attrition index for agglomerates at high tap numbers.

III. MICROSTRUCTURAL APPROACH FOR COMPRESSION AND COMPACTION

A. SCANNING ELECTRON MICROSCOPY STUDIES IN FOOD POWDERS

Because a correlation exists between the microstructure and physical properties of food powders (Aguilera and Stanley, 1999; Kaláb, 1979), the SEM method is appropriate in studying the surface morphology and the internal structure of agglomerated powders. Careful microscopic examination of the failed pieces can provide much insight on the dominant failure modes (Bika *et al.*, 2001). Current micromechanical modeling has clearly demonstrated the importance of structure (packing and resulting porosity) on the properties of agglomerates. However, those models are highly idealized and not capable of predicting quantitatively the mechanical properties of real agglomerates from the properties of constituent materials (Aguilera and Stanley, 1999).

Some agglomerates of different materials have been observed to fail because of internal flaws driven by a number of stresses (e.g., internal tensile stress; cracks in the surface; plastic flow at the surface between the agglomerate and platen; and shear stress within the sphere). For brittle particle agglomerates with significant internal flaws, the tensile strength is small compared to the compressive and shear strength, and failure is likely initiated by the internal tensile stress. In any case, a careful microscopic examination of failed pieces can provide much information on the dominant failure mode (Bika *et al.*, 2001).

Ghadiri *et al.* (1991) used SEM to study the impact damage on NaCl particles developed from different processing routes. Local plastic deformation phenomena leading to cracks, including lateral cracks, and flaw paths are illustrated in Ghadiri *et al.* (1991). Yan and Barbosa-Cánovas (1997)

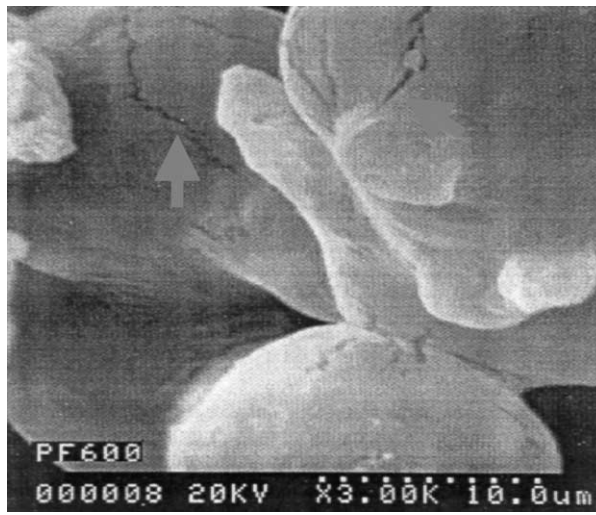

FIG. 19 Fracture lines in a milk powder agglomerate after compression (from Yan and Barbosa-Cánovas, 2000).

studied the microstructure of spray-dried instant low-fat (5%) and nonfat milk before and after compression tests, to characterize their internal forces and compression mechanisms. Before compression, the primary particles in nonfat milk powder were characterized by wrinkled surfaces with deep dents. The 5% low-fat milk powders particles were spherical with small dents and smooth surfaces, with some rims around small globules emerging from the large particles and some craterlike scars. In both cases, the solid bridge force (lactose recrystallization during the instantizing process (Carić and Kaláb, 1987) dominated the overall interparticle forces. Because both had very low moisture contents, the chance for liquid bridge formation was diminished. Very small particles adhering to larger ones could be the result of interparticle attraction forces.

In Figure 19, crack lines can be observed in low-fat milk due to the primary force-transmitting routes of the applied compression stress. As compression proceeds, particle fracture occurs along these crack lines, load paths bifurcate, and more particles become load bearing. Figure 20 shows how large forces tend to be transmitted similarly along particle chains, as observed in experiments on the assembly of photoelastic discs. These particle chains are positioned to form enclosed circular boundaries, where loaded particles create concentrated failure lines and propagate major principal stress.

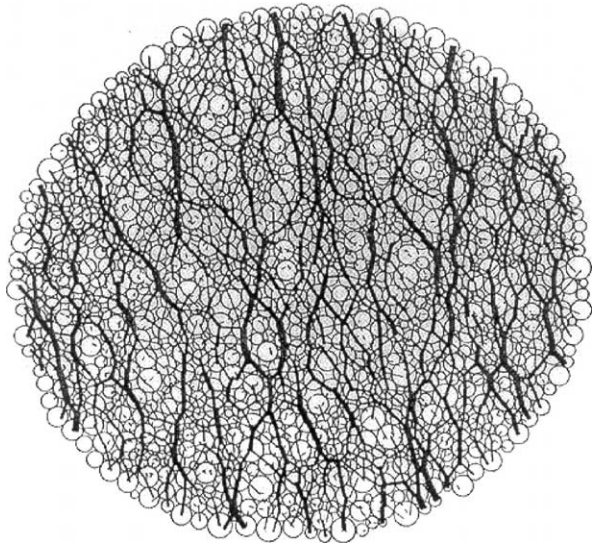

FIG. 20 Force transmission chains through a granular material while under compressive stress (from Yan and Barbosa-Cánovas, 2000). Width of lines represents the relative magnitude of local stress; the principal stress is vertical.

B. FRACTAL CHARACTERIZATION FOR COMPACTION PROCESSES

The fractal approach analyzes two-dimensional projections of particles by redefining its contour. A given step length can provide a close polygonal contour at different points of the particle projection that geometrically recreates a self-similar particle silhouette. The procedure is based on estimating the silhouette perimeter by adding all the step lengths plus the exact length of the last side to complete the polygon (if different from the step length). Measurement of the silhouette perimeter is repeated by using several different step lengths, which gives different perimeters in each case. The perimeter L_P of the corresponding polygon is expressed as (Peleg and Normand, 1985a):

$$L_P = n\lambda, \tag{35}$$

where n is the number of the polygon's sides for a particular selected step length λ. A logarithmic plot of the perimeter L_P against different step lengths λ is called a Richardson plot, which produces a curve with a negative

slope α due to perimeter increase through step length decrease. The fractal dimension D_F of the profile is expressed as

$$D_F = 1 + \tan|\alpha|. \tag{36}$$

The fractal dimension provides useful information about the particle shape, openness, and ruggedness in the form of single numbers (Simons, 1996). Thus, fractal characterization can be used as a tool to describe the eroding process in particulate materials during compaction (Barletta and Barbosa-Cánovas, 1993b). In fact, fractal dimension has been used as a sensitive attrition index, based on the fact attrition can cause changes in particle shape and surface, and on the scale that a fractal approach is applicable (Peleg and Normand, 1985a). Computer algorithms are generally applied for fractal determination using digitized particle images with special software (Allen *et al.*, 1995). Particles with different origins and shapes, such as carbon aggregates, minerals, protein, catalysts, and instant coffee, showed fractal dimensions (D_F) within the range of 1.05–1.36 (Clark, 1986). Particles with D_F close to 2.0 are not likely to exist because of inherent mechanical instability (Normand and Peleg, 1986; Peleg and Normand, 1985b).

Barletta and Barbosa-Cánovas (1993b) used fractal analysis to characterize the changes in ruggedness of particles from attrition by tapping in instant coffee and instant skim milk. Fractal dimension changes in ruggedness were detected even for low tap numbers decreasing with increasing number of taps. It was proposed that fractal analysis be used for quality assessment of instant food powders and other particulate materials for attrition characterization in compaction or compression processes.

IV. COMPRESSION AND COMPACTION IN FOOD PROCESSING

In powder technology, the general behavior of powders under compressive stress or compaction due to mechanical motion is relevant in several applications. The tendency of a food powder's physical and chemical properties to change relative to temperature-moisture history is a common feature of all food powders (Peleg, 1978). Intrinsic variables like temperature, moisture, and composition can influence the response of food powders to the stress of deformation from tension, shear, or compression.

Apart from the influence of temperature and relative humidity conditions during powder processing, different unit operations can provide unique settings for food powder materials to encounter compression and compaction phenomena. In the following sections, we discuss how compaction and compression events directly or indirectly intervene in different processing operations.

A. SIZE REDUCTION

Size reduction operations can be related with particle or powder bulk volume reduction. The objective of particle size reduction is to produce smaller particles from larger ones. Furthermore, bulk powder volume reduction through compression or compaction can also be considered a size-reduction operation.

Compression characteristics are extremely important for bulk size–reduction operations (Murthy and Battacharya, 1998), especially in military and domestic applications. Compressed powders can be used by armed forces for field rations, by astronauts during space travel, and by hikers and others who must carry their own food supplies. Powders can be compressed by mechanical presses with savings in transportation costs, storage space (Van Heyst, 1983), and packaging material. Webb and Hufnagel (1943) reported a 42% savings in space with compressed dry whole milk (Van Heyst, 1983).

Particle size reduction, a comminution process, includes operations such as crushing, grinding, and milling (e.g., milling of cereals, grinding of spices) in which food pieces are deformed until breakage or failure. As mentioned earlier, breakage can occur along cracks or defects in the structure of hard materials during compression. Scanlon and Lamb (1995) showed how different types of fracture operations affected the particle shape differently in dried gelatinized starch comminuted in an impact breakage gun, hammer mill, and blender.

Forces commonly used in food processes for particle size reduction are compressive, impact, attrition (or shear), and cutting forces. More than one force usually participates in the comminution operation in industrial size-reduction equipment. In particular, crushing rolls use mainly compressive forces, hammer mills are based on impact, disc mills cause particle attrition through shear force application, and rotary knife cutters use cutting forces.

Coarse crushing of hard materials through compressive forces allows a solids reduction to about 3 mm. Impact forces may be associated with coarse, medium, and fine grinding for a variety of food materials (e.g., for nut breakage). Shear or attrition forces are applied for pulverization of powders in the micrometer range (like most food powders). Cutting is a process totally different from comminution, because the operating principles are quite different from those governing the size reduction of hard materials and generally gives a definite particle size and may even produce a definite shape. An ideal size-reduction pattern to achieve a high reduction ratio for hard brittle materials, such as sugar crystals or dry grains, could be obtained by first compressing and then using an impact force, and finally by shearing or rubbing.

In regards to particle shape and defects, a large piece having many defects can be broken under small stress with very little deformation. On the other hand, smaller pieces have fewer defects and will need a higher breaking strength. Limited by very small particles, purely intermolecular forces must be overcome. This is why grinding is so difficult to achieve in smaller particles. For example, fine grinding of roasted coffee (e.g., to <50 μm) is best recommended under cryogenic conditions (i.e., subzero temperatures) to accomplish the desired grinding efficiency. Coarse crushers have size-reduction ratios of less than 8:1, whereas fine grinders have ratios as high as 100:1. However, large reduction ratios, such as those obtained when grinding relatively large solid lumps into ultrafine powders, are normally attained in several stages using diverse crushing and grinding machines. A good example of this is the overall milling of wheat grain into fine flour, in which crushing rolls in a series of decreasing diameters are employed.

Knowledge of the compression properties of feed materials can indicate the type of force most likely needed in size reduction. A friable or crystalline structure can be reduced through fracture along cleavage planes using compressive forces. However, if new crack tips must be formed, impact and shear forces may be more effective. For food materials of fibrous structures, shredding or cutting should be considered for the desired size reduction.

Hard brittle materials like sugar crystals can be crushed, broken by impact, or ground by abrasion. For example, recognition of the different grinding properties of sugar and cocoa powder is resulting in a change in the grinding procedures (Niediek, 1971). The traditional process is to grind the sugar–cocoa mixture together, but because sugar is brittle and cocoa is ductile, it is better to grind the sugar by pressing (e.g., in a traditional mill) and the cocoa separately by impact (e.g., in a hammer mill) (Loncin and Merson, 1979).

B. SIZE ENLARGEMENT

Size enlargement involves two main types of operations: (1) agglomeration (or compaction, granulation, tabletting, briquetting, pelletizing, sintering) and (2) encapsulation, which combines smaller particles into larger composites of identifiable unit components. Applications in food processing are numerous and becoming increasingly important as more structured foods are developed. Some of these processes include compression and compaction during compact formation, but other processes are needed that are designed so that properties of final products will resist mechanical attrition during handling.

1. Agglomeration

Agglomeration is a process aimed at controlling particle porosity and density. The aggregation of dispersed materials into materials with larger unit size, held together by adhesive and/or cohesive forces, is known as *agglomeration* (Bika *et al.*, 2001). There are two main types of agglomeration: *rewetting* and *pressure*.

In rewetting agglomeration, the surface of the dried particles is rewetted with steam or water misting, and particles are then mixed usually in a turbulent gas stream, which causes them to form clusters by collisions. Then the agglomerates are re-dried and sized. In general, agglomerates have a coarse open structure of about 100–3000 μm. Some agglomeration applications are milk powder, spray-dried coffee, flours, starches, dry soups, cocoa products, dextrins, and dry pudding mixes production (Hall and Hedrick, 1971). Rewetting agglomeration is used in food processes partly to improve properties related to handling.

Pressure agglomeration is the most relevant agglomerated process for compaction studies because particulates are confined by compression into a mass that is then shaped and densified. Two compacting pressures of different magnitudes are applied for preset periods of time: (1) a low pressure for particle rearrangement and (2) a pressure high enough to break brittle particles uniformly and to plastically deform malleable particles (Pietsch, 1994). The feed mixture is often prepared with fine particles and binders, thus giving a sticky mass, which may be formed by forcing it through holes in differently shaped screens or perforated dies in extruders (Halliday and Smith, 1997) or presses. Agglomeration and shaping are, therefore, due to pressure forcing the material through the holes, as well as frictional forces.

One particular example of pressure agglomeration (Pietsch, 1999) is the compact/granulation process whereby dry powdered mixtures are first compacted by high pressure, and then crushed and screened into a granular (instant) product. Another example is briquetting and tableting whereby compaction of food ingredients such as dextrose, gelatin, glucose, sucrose, lactose, and starch are pressed with food gums as binders. Pressure compaction with binders in roll or plate presses or pellet machines is used for candies and dried soups.

a. Strength of final agglomerates. Bonds that connect particles to form granules must be sufficiently strong to prevent breakdown of the final dried granules to powder in subsequent handling operations. Bond strength is determined by the particle size, the structure of the granule, moisture content, and surface tension of the liquid, as well as the presence of solid

bridges, liquid bridges (immobile or freely movable), Van der Waals forces, electrostatic forces, and interlocking bonds (Parikh, 1997).

In general, adhesive forces increase linearly with particle size. Once agglomerates are formed, the strength depends largely on liquid bridges if moisture is present; otherwise, the weaker Van der Waals forces are important. Agglomerates are inherently multiphase materials, with at least one fluid phase contained in the interstitial volume between primary particles. The force of cohesion between solid bridges in dried agglomerates depends on the diameter of the contact area and the strength of the bridge material, which can be hardened by binder substances. One exception to the rule is when the material is a charge insulator; in this case, attraction forces vary with the square of the diameter.

Although the strength of single agglomerates depends on the forces holding the agglomerate together, it is often difficult to calculate the strength of agglomerates based on any one type of force. One possibility is to express the strength of agglomerates as the tensile strength σ_t necessary to hold component particles together.

$$\sigma_t = \frac{1-\epsilon}{\epsilon}\frac{\sum_{i=1}^{n} A_i(x,\ldots)}{x^2}, \tag{37}$$

where A_i is the adhesion force caused by a particular binding mechanism and x is the representative size of the particles forming the agglomerate. A_i is also a function of other unknown parameters. Other models are available for predicting adhesion forces of various types (Pietsch, 1991), but A_i changes its magnitude for each bond because of differences in roughness of each particle forming the agglomerates. Food agglomerates have lower mechanical strength than inorganic or polymeric particulates (Bemrose and Bridgwater, 1987). Moreover, agglomerates have better flowability (or less cohesion) when obtained from larger size particles (Schubert, 1981, 1987).

Food agglomerates obtained from rewetting are normally brittle and easily broken when exposed to mechanical impact or vibration during processing and transportation (Barletta *et al.*, 1993a). Collisions among brittle agglomerates and against the static container walls due to mechanical compression or vibration easily provide the kinetic energy necessary to cause both attrition and compaction (Barletta and Barbosa-Cánovas, 1993a). On the other hand, products from high-pressure agglomeration possess high strength immediately after discharge from the equipment. Addition of small amounts of binders or use of posttreatment methods can increase agglomerate strength even more.

b. Strength of green agglomerates. In pressure agglomeration, particle strength not only is relevant for the final product but also plays a role during the size enlargement operation. Green (or wet) agglomerates are formed first and then must be cured (bonds must be stabilized by compression) to obtain permanent bonding. For green agglomerate binding stability, matrix binders and high temperatures can help strengthen bonding. The stresses such food powders develop in storage are usually a few orders of magnitude smaller than stresses developed in tableting or similar operations (Peleg, 1983). Agglomerates formed by rewetting agglomeration attain lower strength levels primarily because they feature higher porosity from coalescence, whereas pressure agglomeration causes porosity to decrease while density and strength increase.

2. *Encapsulated materials resistance*

Encapsulation is a process in which a continuous thin coating is formed around solid particles (e.g., powdered sweeteners, vitamins, minerals, preservatives, antioxidants, cross-linking agents, leavening agents, colorants, and nutrients) to create a capsule wall (King, 1995; Risch, 1995). Encapsulation promotes easier handling of the core or interior material by preventing lumping, by improving flowability, compression, and mixing properties, and by reducing core particle dustiness and modifying particle density (Shahidi and Han, 1993).

The capsule structure is divided into two parts: (1) the core, containing the interior contents and (2) the coating material (e.g., gums). One cannot generalize about compaction behavior of microcapsules because diverse structures exist. Microcapsules can be classified, however, into three main structures: (1) single particle (regular or irregular), (2) aggregate, and (3) multiwalled. In particular, multiwall structured capsules contain different concentric layers of the same or different composition and earn greater resistance to attrition during handling. Microcapsules contain and protect the core material inside the shell during storage, providing protection from oxidative deterioration and avoiding active nutrients lost (Imagi *et al.*, 1992; Onwulata *et al.*, 1998). However, the coating must be developed in such a way that it can be fractured by external forces, such as pressure, shearing, or extrasonics in the range of compressive and impact forces produced during mastication.

An effective coating material should have good rheological properties at high concentration. Viscosity, thermal stability, solubility/meltability, and film-forming ability of a coating material are critical for its final strength. Furthermore, coating materials must be easy to manipulate during the process of encapsulation. For example, during air suspension coating using

fluidized bed technology, particle shape and size (ranging between 50 and 500 μm) will depend on how the coating has been attached. Both factors are critical to the final quality of the encapsulated product. The more spherical the particle is, the better its encapsulation will be because if sharp edges protrude through the applied coating surface, the capsule can become vulnerable to release (DeZarn, 1995).

Microencapsulation by spray drying of fats reduces adhesiveness (reducing clumping and caking) and enhances handling properties during storage transport and blending with nonfat ingredients. Such powders should resist compaction forces that could possibly rupture the capsule during packaging, shipping, and storage, because desirable flow characteristics are impaired by encapsulated fat on particle surface.

C. MIXING, HANDLING, AND TRANSPORTING

Mechanical damage to powdered foods usually results from compressive loads (Mohsenin, 1986). Physical properties of agricultural food materials are important in postharvest unit operations for the design of storage structures and for selecting the handling equipment (Murthy and Battacharya, 1998). Particularly, compaction, conveying, mixing, and metering among other types of food powder handling can provoke attrition (Schubert, 1987), bringing problems such as changes in bulk properties, segregation, and in some agglomerates, loss of instantaneity.

1. Compaction during mixing

Compaction during mixing occurs when free or easy-flow powders with significant ranges in particle size are exposed to gravitational, rotational, vibratory, or aeration operations or other types of mechanical motion. Different compaction dynamics are observed during blending, where finer or less coarse particles congregate in the center of the rotating device when angular speed is low. For example when tumbling or stirring, if particles are coarse enough, segregation mechanisms will occur in the sliding layers on sloping surfaces continuously created in the equipment, thus decreasing mixing efficiency (Enstad, 2001). Mathematical modeling of different mixing processes and compaction phenomena such as food particle segregation or particle resistance to attrition or erosion is scarce, which makes it difficult to develop relationships between mixing and quality (Niranjan, 1995), especially when blending food powders due to the fragile nature and different sizes of food products (Niranjan and de Alwis, 1993). Powder bulk properties, like cohesiveness and stickiness, make food particulate mixing a complicated operation.

Mixing mechanisms can be affected by compaction properties such as mechanical interlocking, surface attraction, plastic welding (from high pressures between small contact areas), electrostatic attraction, and environmental factors (e.g., ambient moisture and temperature fluctuations). Powder flow properties can simplify blender selection by describing the behavior of materials with specific compositions in different types of mixers, which takes into consideration there are no stagnant regions (or areas where materials can settle undisturbed separate from the mixing process) in the blender, and that different flow velocities in various sections of the blender are promoted and blender segregate demixing is avoided (Dudley, 2001). In many cases, these conditions depend on bulk properties like cohesiveness and angle of repose, which may change with product formulation. Dudley (2001) provided a table where mixers are classified in different angle of repose ranges.

Kuakpetoon *et al.* (2001) studied the effect of particle size, shape, surface, and mixing ratio on the characteristics of dry flour mixes, using a laboratory drum mixer and a double-ribbon mixer. Flour mixtures with smaller size particles (5–50 μm, spherical-oval shapes, and smooth surfaces) achieved high uniformity (or standard deviation) but required a longer mixing time. In contrast, mixes with larger sizes (50–150 μm, irregular shapes, and very rough surfaces) had a low degree of mixing but required a shorter time to reach uniformity. Differences in angle of repose, tensile strength, and true density measurements were also observed.

2. Compaction during conveying

Different conveyors such as the belt, chain, and screw types, as well as pneumatic equipment, are used to transport bulk powdered foods. Conveyor belts are used for movement of different types of bulk solids at long distances. The belt and its load are supported on idlers on both conveying and return sections. The material can be discharged over the end of the belt either by using a diagonal scraper, by tilting one or more of the idler pulleys, or by using a tripper.

When a bulk solid is loaded onto a conveyor belt, it is loosely packed with a surcharge angle approximating the static angle of repose (Roberts, 2001). However, the material will soon return to its equilibrium packing condition due to the motion over the idlers, and segregation will occur within the bulk materials with fines and moisture will migrate to the lower belt surface. The carrying capacity depends on the cross-sectional area of the material and the belt, and the load profile depends on the method of loading and the properties of the bulk solid. During transport, the packing density ratio (i.e., ratio between bulk and solid density) approaches an asymptotic value, as predicted by Equation 22 and other packing models discussed. When the

powder is dropped on the belt, its load point corresponds to the maximum consolidation stress $\sigma_{1\text{discharge}}$.

$$\sigma_{1\text{discharge}} = k_L \rho_b g h, \tag{38}$$

where h is the average depth of bulk solid on the conveyor belt at the load point, ρ_b is the bulk density at the load point corresponding to σ_1 discharge, and k_L is the load factor (generally ranging between 0.5 and 1.0). If the powder is dispensed on an inclined belt, there will be reduced friction between the bulk solid and the belt, leading to slippage during inclined conveying (Roberts, 1998).

D. PACKAGING: COMPRESSIBILITY USING PADDING MATERIALS

Some cushioning materials in the powder bed or on the container wall can absorb or reduce the mechanical or static impact on the agglomerates, which reduces their degree of attrition. Using the confined uniaxial compression test, Yan and Barbosa-Cánovas (2001b) studied the padding effect on agglomerated coffee and nonfat milk using pure polyurethane foam as padding material. The padding effects were evaluated by using the proposed padding index I_p and padding efficiency E_p. The padding index I_p is defined as

$$I_p = 1 - \frac{d_1 - H}{d_0} (0 < I_p < \infty), \tag{39}$$

where d_1 is the final deformation of powder bed when padding material is used, d_0 is the final deformation of the powder bed without padding, and H is the initial thickness of the padding material. A higher padding index indicates that much more deformation is induced in the padding material instead of the powder bed. The padding efficiency E_p is defined as

$$E_p = \frac{f_1 - f_0}{f_0} \times 100\% (0 < E_p < \infty), \tag{40}$$

where f_1 is the weight fraction of agglomerates retaining their original particle size when padding material is used, and f_0 is the weight fraction of agglomerates retaining their original particle size without padding. Padding efficiency is more meaningful than the padding index for agglomerated food powders in expressing the effects of padding application on reducing attrition. Yan and Barbosa-Cánovas (2001b) conducted tests under four conditions: without padding, and with padding foams (polyurethane, 7 and 13 mm thick) at the cell bottom, at the top, and at the middle (Figure 21). The compressive force-deformation relationship of polyurethane foam had a characteristic exponential shape, as shown in Figure 22.

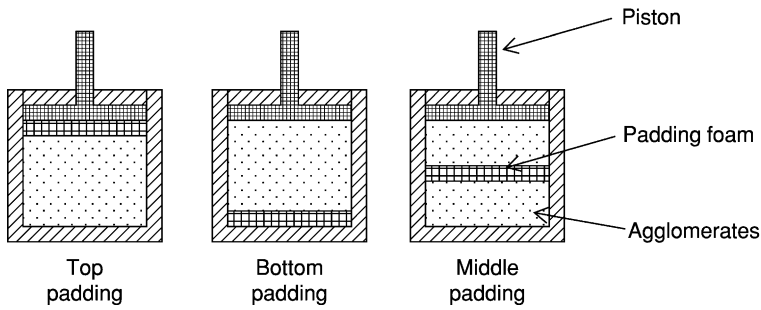

FIG. 21 Three positions for padding foam in the powder bed, as confined in compression cells (from Yan and Barbosa-Cánovas, 2000).

The force-deformation curves were smoother for instant coffee when padding was used. However, curves for instant milk retained an exponential characteristic. A change in the foam position caused differences in the overall deformation. Both padding index and efficiency for both powders were at peak values when padding material was at the top. As more deformation was induced in the padding material (i.e., more padding index), less deformation was induced in the powder bed (i.e., more padding efficiency), so fewer particles were damaged.

The padding efficiency was better for lower strength milk agglomerates than for harder strength agglomerates such as coffee. The thickness of the polyurethane made no significant difference in the padding values. The padding index was sensitive to thickness differences but not nearly as sensitive to different padding positions, mainly due to the unavoidable random initial packing of the agglomerates in the compression cell.

E. BULK STORAGE

Compaction of food powders enables a reduction in bulk storage, allowing more efficient mechanical handling of powders in smaller storage spaces and easier transportation (Mohsenin, 1986). Furthermore, the study of stresses developed as a result on the storage of powders in high bins, hoppers, or silos has played a key role in compression properties evaluation. Because bulk density is one of the most important characteristics of bulk powder storage operations, mechanical compressibility can provide an idea of bulk density changes due to compacting pressure in stored powders. Different high-pressure operations and other problems caused by hydrostatic compression and compaction during storage and discharge are discussed in the following sections.

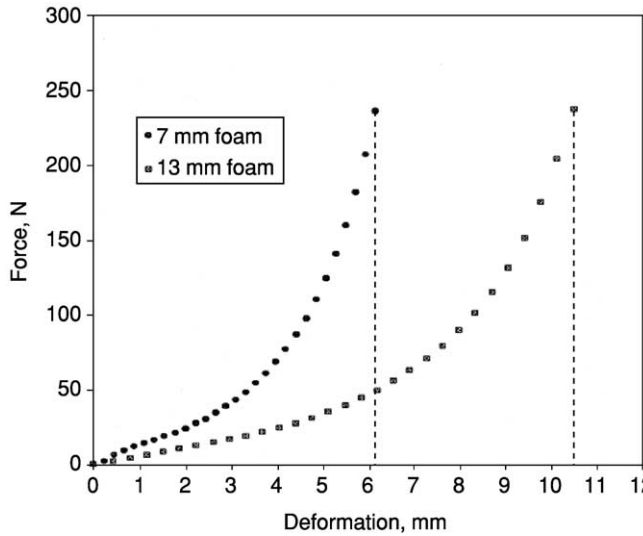

FIG. 22 Force-deformation relationship for two padding foams of different thicknesses (from Yan and Barbosa-Cánovas, 2000).

1. *Bulk storage reduction*

Cenkowski *et al.* (2000) studied the storage volume reduction of flour by mechanical compression as a way of offering advantages for long-term storage. By reducing storage volume to 55%, oxygen diffusion into the flour was slowed down; therefore, storage stability was improved 5–15% by reducing oxidative processes. Compacted flour also offers the advantage of making flour more resistant to possible infestation by mites or other microorganisms. The economics of compacting milled wheat has been investigated by Hassan *et al.* (1973) at hydrostatic pressures up to 4.1 MPa. They concluded that substantial savings could be achieved in the storage and handling costs of compacted agricultural granular materials. Furthermore, Loewer *et al.* (1977) studied the low-pressure compaction of ground corn (7–70 kPa) as it relates to forces in bulk storage structures.

2. *Compression phenomena in bins and feeders*

A feeder usually consists of a vertical part called the *bin* and a converging part called the *hopper*. In the storage bin, the bulk solids normal stress increases linearly with depth due to the weight of individual particles while they transmit static shear forces. Stress at the wall quickly reaches a

maximum value as depth increases, because part of the bulk solids weight is transmitted to the walls via friction forces. In the hopper, the vertical pressure is decreasing, because in addition to the friction force between the bulk solid and the wall, the vertical component of the normal stress at the wall carries part of the weight of the bulk solid. If the bulk solid is discharged through the bottom opening, the material in the hopper is compressed horizontally—due to the converging action—while it expands vertically due to the open outlet.

3. Arch and rathole formation

Arches and ratholes, both caused by compression phenomena, are the most common flow obstruction problems caused by powdered material stored in hoppers and bins. An arch is a stable obstruction that generally forms over the discharge outlet, which supports the rest of the bin's contents, preventing discharge. Ratholes (or pipes) are bulk solid materials found above the outlet, which remain stagnant in dead zones of bins and hoppers. Cohesive strength causes this stagnant material to bind together or interlock, forming a narrow channel above the outlet where material can flow and discharge, and decreasing the usable capacity of the bin. Because rathole material remains under storage, it can cake or degrade.

The unconfined yield strength of a bulk solid is the main property associated with arching and rathole formation. In fine or sticky powders, arching can result from cohesive forces, while in the case of coarse bulk solids, arching is caused by either the interlocking or the frictional force of single particles. Consolidation time is crucial in both cases, especially in rathole formation. A rathole can collapse if the stress imposed on the material exceeds its yield strength.

Compressibility values can be used to determine the minimum dimensions required to overcome arching and ratholing, by estimating the bulk density of the material at the outlet of the hopper. Furthermore, the loads acting on a feeder gate will depend on the bulk density of the solid at the point where the feeder or gate is located (Thomson, 1997).

4. Segregation in bulk storage

One common compaction mechanism is segregation. During powder storage in bins and hoppers, segregation can be significant when there is a horizontal shear movement of particles on a free pile surface. The powder heap has a predominance of fines toward the center of the bin because of sifting during bin discharging. An example of an obvious segregation problem is a drink mix that varies in tartness due to fluctuations in the citric acid content. A less

obvious example would be packages that are routinely overfilled to ensure they meet the weights stated on the label.

V. CONCLUSION

Food powder compression is involved in different industrial applications such as bulk size reduction, grinding, particle size enlargement, food tablet production, encapsulated material resistance, hopper and silos storage design, mixing, and packaging. Handling operations are related to mechanical properties of materials (shear stress, tensile, and compressive stress), in that brittle agglomerated food powders can suffer the undesirable effect of particle breakage or attrition.

Different commonly used compression characterization methods have been demonstrated: the "Brazilian test," the confined uniaxial and unconfined compression test, and the flexible boundary cubical triaxial test. The HHP method appears as a new and promising possibility within the traditional compression methods. In HHP, higher pressure gives higher bulk density; however, beyond a critical given pressure, the final compressed bulk density remains constant (ultimate bulk density). Further studies will determine whether this concept can be included as a quality descriptor in the specification data sheets of commercial food agglomerates.

Compression models have been introduced to characterize food powder compression mechanisms. The Heckel equation describes the densification phenomenon for a first-order compaction kinetics. Sone's model is a simple form that can describe certain physical properties such as compressibility and flowability for fine and agglomerated powders in compression tests, although compressive pressure range limits exist for its application. The Swyngedau's four-parameter model has proven successful in describing the sigmoid-shaped force-deformation relationships; its parameters provide numerical values that can describe the detailed compressive behavior of agglomerates during different steps of compression. The compression characteristics, measured as the compressibility of food powders are influenced by particle size, mixture composition, water activity, and compression cell geometry and can be assessed based on the selection of anticaking agents by describing a powder's cohesion. Compressibility tests give an approximate measure of powder flowability. It is not suitable for the design of silos but may be a convenient method for process control.

The microstructure of broken-down agglomerates and fractured primary particles due to compression can be studied by SEM. SEM observations of agglomerate microstructure after compression can provide detailed visual description of the behavior of milk powder with insight into the compression

mechanisms. Crack lines in agglomerated powders give useful information on the force transmission patterns in the compressed bed.

The force-deformation relation and the fraction of particles retaining original particle size after compression of selected food agglomerates can be affected by padding foams. The padding index Ip and padding efficiency Ep values showed that the padding effect is more obvious when padding material is placed at the top of the powder bed. Furthermore, the padding foam thickness makes no significant difference and the padding material is more beneficial in lowering the internal strength of agglomerates.

Relevant topics that need to be addressed are the development of improved padding materials for conveying and bulk storage, further microstructure analysis of encapsulated materials, the potential use of highly compressed powders in the food industry, to keep working in developing a comprehensive data base on the bulk powder volume reduction during storage with respect to composition and variation in relative humidity, temperature, and particle size. In addition, the identification of key forces (e.g., shear, frictional, and structural) most likely needed in size reduction of different feed materials can be of help in the development of size reduction equipment. In this line, nanotechnology is bringing ultraprecision machine systems that will allow researchers to better understand processes and functional properties of particulate materials from the molecular perspective.

REFERENCES

Adams, M.J., Mullier, M.A., and Seville, J.P.K. 1994. Agglomerate strength measurement using uniaxial compression test. *Powder Technol.* **78**, 5–13.

Adhikari, B., Howes, T., Bhandari, B.R., and Truong, V. 2001. Stickiness in foods: A review of mechanisms and test methods. *Int. J. Food Prop.* **4**, 1–33.

Aguilera, J.M. and Stanley, D.W. 1999. "Microstructural Principles of Food Processing and Engineering". Aspen Publishing, Gaithersburg, MD.

Allen, M., Brown, G.J., and Miles, N.J. 1995. Measurement of boundary fractal dimensions: Review of current techniques. *Powder Technol.* **84**, 1–14.

American Society for Testing and Materials 1986. "Compilation of ASTM standard definitions". American Society for Testing and Materials, Baltimore, MD.

American Society for Testing and Materials 1997. Standard shear testing method for bulk solids using the Jenike shear cell. American Society for Testing and Materials, Baltimore, MD.

Barbosa-Cánovas, G.V. and Juliano, P. 2005. Physical and Chemical Properties of Food Powders. Chapter 2. *In* "Food Powders and Encapsulated" (R.P. Onwulata and C. Konstance, eds). In Press.

Barbosa-Cánovas, G.V., Malavé-López, J., and Peleg, M. 1987. Density and compressibility of selected food mixtures. *J. Food Eng.* **10**, 1–19.

Barbosa-Cánovas, G.V., Malavé-López, J., and Peleg, M. 1985. Segregation in food powders. *Biotechnol. Prog.* **1**, 140–146.

Barbosa-Cánovas, G.V. and Yan, H. 2003. Powder characteristics of preprocessed cereal flours. *In* "Characterization of Cereals and Flours: Properties, Analysis, and Applications" (G. Kaletunç and K.J. Breslauer, eds). Marcel Dekker, New York.

Barletta, B.J. and Barbosa-Cánovas, G.V. 1993a. An attrition index to assess fines formation and particle size reduction in tapped agglomerated food powders. *Powder Technol.* **77**, 89–93.

Barletta, B.J. and Barbosa-Cánovas, G.V. 1993b. Fractal analysis to characterize ruggedness changes in tapped agglomerated food powders. *J. Food Sci.* **58**, 1030–1046.

Barletta, B.J., Knight, K.M., and Barbosa-Cánovas, G.V. 1993a. Compaction characteristics of agglomerated coffee during tapping. *J. Texture Stud.* **24**, 253–268.

Barletta, B.J., Knight, K.M., and Barbosa-Cánovas, G.V. 1993b. Review: Attrition in agglomerated coffee. *Rev. Esp. Cienc. Tecnol. Aliment.* **33**, 43–58.

Bell, T.A. 2001. Solids flowability measurement and interpretation in industry. *In* "Handbook of Conveying and Handling of Particulate Solids. Handbook of Powder Technology" (A. Levy and H. Kalman, eds), pp. 3–14. Elsevier Science, Amsterdam, The Netherlands.

Bemrose, C.R. and Bridgwater, J. 1987. A review of attrition and attrition test methods. *Powder Technol.* **49**, 97–126.

Benbow, J.J. 1983. Mechanism of compaction. *In* "Enlargement and Compaction of Particulate Solids" (N.G. Stanley-Wood, ed.), p. 161. Butterworth, London, UK.

Bika, D.F., Gentzler, M., and Michaels, J.N. 2001. Mechanical properties of agglomerates. *Powder Technol.* **117**, 98–112.

Biscans, B., Chemini, R., Guiraud, P., and Laguerie, C. 1996. Design of an attrition experiment to simulate the effects of crystal-wall or crystal-stirrer impacts occurring in a crystallizer. *Powder Technol.* **86**, 155–161.

Buma, T.J. 1971. Free fat in spray-dried whole milk 5. Cohesion: Determination, influence of particle size, moisture content and free-fat content. *Netherlands Milk Dairy J.* **25**, 107–122.

Carić, M. 1994. "Concentrated and Dried Dairy Products". VCH Publishers, New York.

Carić, M. and Kaláb, M. 1987. Effects of drying techniques on milk powders quality and microstructure: A review. *Food Microstruct.* **6**, 171–180.

Carson, J.W. and Pittenger, B.H. 1998. Bulk properties of powders. *In* "Powder Metal Technologies and Applications" (R.J. Davis, ed.), pp. 287–301. ASM International Handbook Committee, ASM International, Materials Park, OH.

Carr, E. 1965. Evaluating flow properties of solids. *Chem. Eng.* **72**, 163–168.

Carr, R.L. 1976. Powder and granule properties and mechanics. *In* "Gas-Solids Handling in the Processing Industries" (J.M. Marchello and A. Gomezplata, eds). Marcel Dekker, New York.

Cartensen, J.T. and Hou, X.P. 1985. The Athy-Heckel equation applied to granular agglomerates of basic tricalcium phosphate [$3Ca_3PO_4.Ca(OH)_2$]. *Powder Technol.* **42**, 153–157.

Cenkowski, S., Dexter, J.E., and Scanlon, M.G. 2000. Mechanical compaction of flour: The effect of storage temperature on dough rheological properties. *Can. Agric. Engin.* **42**, 33–41.

Chang, K.S., Kim, D.W., Kim, S.S., and Jung, M.Y. 1998. Bulk flow properties of model food powder at different water activity. *Int. J. Food Prop.* **1**, 45–55.

Chen, M. and Malgham, S.G. 1994. Investigation of compaction equation for powder. *Powder Technol.* **81**, 75–81.

Clark, N.N. 1986. Three techniques for implementing digital fractal analysis of particle shape. *Powder Technol.* **46**, 45–52.

Couroyer, C., Ning, Z., and Ghadiri, M. 2000. Distinct element analysis of bulk crushing: Effect of properties and loading rate. *Powder Technol.* **109**, 241–254.

Denby, J. 1973. Isostatic pressing. *In* "Particular Matter", Special POWTECH '73. Issue 1973.

DeZarn, T.J. 1995. Food ingredients encapsulation. *In* "Encapsulation and Controlled Release of Food Ingredients" (S.J. Risch and G.A. Reineccius, eds), pp. 75–86. American Chemical Society, Washington, DC.

Duberg, M. and Nyström, C. 1986. Studies of direct compression of tablets. XVII. Porosity-pressure curves for characterization of volume reduction mechanisms in powder compression. *Powder Technol.* **46**, 67–75.

Dudley, L. 2001. Choosing a blender based on your material's flow properties. *Powder Bulk Eng.* **15**, 37–39.

Ehlermann, D.A.E. and Schubert, H. 1987. Compressibility characteristics of food powders: Characterizing the flowability of food powders by compression tests. *In* "Physical Properties of Foods—2" (R. Jowitt, F. Escher, M. Kent, B. McKenna, and M. Roques, eds). Cost 90bis Final Seminar Proceedings. ECSC, EEC, EAEC, Brussels and Luxembourg.

Enstad, G.G. 2001. Segregation of powders: Mechanisms, process and counteraction. *In* "Handbook of Conveying and Handling of Particulate Solids" (A. Levi and K. Kalman, eds), pp. 589–603. Elsevier, Amsterdam.

Ferdinand, J.M., Kirby, A.R., and Smith, A.C. 1990. The compaction properties of dehydrated potato. *J. Food Process Eng.* **12**, 99–112.

Geoffroy, J.M. and Cartensen, J.T. 1991. Effects of measurement methods on the properties of materials. *Powder Technol.* **68**, 91–96.

Georget, D.M.R., Parker, R., and Smith, A.C. 1994. A study of the effects of water content on the behavior of breakfast cereal flakes. *Powder Technol.* **81**, 189–195.

Gerhards, C., Ulbricht, D.M., and Peleg, M. 1998. Mechanical characterization of individual instant coffee agglomerates. *J. Food Sci.* **63**, 140–142.

Gerritsen, A.H. and Stemerding, S. 1980. Crackling of powdered materials during moderate compression. *Powder Technol.* **27**, 183–188.

Ghadiri, M., Yuregir, K.R., Pollock, H.M., Ross, J.D.J., and Rolfe, N. 1991. Influence of processing conditions on attrition of NaCl crystals. *Powder Technol.* **65**, 311–320.

Ghadiri, M. and Ning, Z. 1997. Effect of shear strain rate on attrition of particulate solids in a shear cell. *In* "Powders & Grains 97, Proceedings of the International Conference on Powders & Grains", Durham, NC.

Ghadiri, M. 1997. Particle impact and attrition. *In* "Powder Technology Handbook" (K. Gotoh, H. Masuda, and K. Higashitani, eds), pp. 183–191. Marcel Dekker, New York.

Grey, R. and Beddow, J. 1969. On the Hausner ratio and its relationship to some properties of metal powders. *Powder Technol.* **2**, 323–326.

Hall, C.W. and Hedrick, T.I. 1971. "Drying of Milk and Milk Products". AVI, Westport, CT.

Halliday, P.J. and Smith, A.C. 1997. Compaction and flow of potato starch and potato granules. *Food Sci. Technol. Int.* **3**, 333–342.

Hamano, M. and Sugimoto, H. 1978. Water sorption, reduction of caking and improvement of free-flowingness of powdered soy sauce and miso. *J. Food Process. Pres.* **2**, 185–196.

Hartley, P.A., Parfitt, G.D., and Pollack, L.B. 1985. The role of Van der Waals force in agglomeration of food powders containing submicron particles. *Powder Technol.* **42**, 35–46.

Hassan, Z.A., Finley, R.M., and Johnson, S.R. 1973. The demand for food in the United States. *Appl. Econ.* **5**, 233–248.

Hayes, G.D. 1987. "Food Engineering Data Handbook". John Wiley and Sons, New York.

Head, K.H. 1982. "Manual of Soil Laboratory Testing". Pentech Press, London, UK.

Hollenbach, A., Peleg, M., and Rufner, R. 1982. Effect of four anticaking agents on the bulk characteristics of ground sugar. *J. Food Sci.* **47**, 538–544.

Hollman, J.P. 2001. "Experimental Methods for Engineers". McGraw-Hill, New York.

Imagi, J. Muraya, K., Yamashita, D., Adachi, S., and Matsuno, R. 1992. Retarded oxidation of liquid lipids encapsulated in matrixes of saccharides or proteins. *Biosci. Biotech. Biochem.* **56**, 1236–1240.

Kaláb, M. 1979. Microstructure of dairy foods. Milk products based on protein. *J. Dairy Sci.* **62**, 1352–1364.

Kamath, S. 1996. "Constitutive parameter determination for food powders using triaxial and finite element analysis of incipient flow from hopper bins". Doctoral Thesis, Pennsylvania State University, PA.

Kamath, S., Puri, L., Manbeck, H.B., and Hogg, R. 1993. Flow properties of powders using four testers – measurement, comparison and assessment. *Powder Technol.* **76**, 227–289.

King, A.H. 1995. Encapsulation of food ingredients. *In* "Encapsulation and Controlled Release of Food Ingredients" (S.J. Risch and G.A. Reineccius, eds), pp. 26–39. American Chemical Society, Washington, DC.

Konstance, R.P., Onwulata, C.I., and Holsinger, V.H. 1995. Flow properties of spray-dried encapsulated butteroil. *J. Food Sci.* **60**, 841–844.

Kuakpetoon, D., Flores, R.A., and Milliken, G.A. 2001. Dry mixing of wheat flours: Effect of particle properties and blending ratio. *Lebensm.-Wiss. U. Technol.* **34**, 183–193.

Kumar, M. 1973. Compaction behavior of ground corn. *J. Food Sci.* **38**, 877–878.

Kurup, T.R.R. and Pipel, N. 1978. Compression characteristics of pharmaceutical powder mixtures. *Powder Tehcnol.* **19**, 147–155.

Lai, C.C., Gilbert, S.G., and Mannheim, C.H. 1986. Effect of flow conditioners on water sorption and flow properties of egg powder. *J. Food Eng.* **5**, 321–333.

Lambe, T.W. and Whitman, R.V. 1969. "Soil Mechanics". John Wiley & Sons, New York.

Li, F. and Puri, V.M. 1996. Measurement of anisotropic behavior of dry cohesive and cohesionless powders using a cubical triaxial tester. *Powder Technol.* **89**, 197–207.

Lindley, L.T. 1991. Mixing processes for agricultural and food materials: 3. Powders and particulates. *J. Agric. Eng. Res.* **49**, 1–19.

Lloyd, P.J. 1983. Mechanism of size enlargement. *In* "Enlargement and Compaction of Particulate Solids" (N.G. Stanley-Wood, ed.), p. 128. Butterworth, London, UK.

Loewer, O.J., Ross, I.J., Kratzer, D.D., and Walker, J.N. 1977. Properties of ground shelled corn as related to forces in bulk storage structures. *Trans ASAE* **20**, 155–156.

Loncin, M. and Merson, R.L. 1979. "Food Engineering. Principles and Selected Applications". Academic Press, New York.

Lu, W.M., Tung, K.L., Hung, S.M., Shiau, J.S., and Hwang, K.J. 2001. Compression of deformable gel particles. *Powder Technol.* **116**, 1–12.

Macleod, H.M. 1983. Compaction of ceramics. *In* "Enlargement and Compaction of Particulate Solids" (N.G. Stanley-Wood, ed.), p. 241. Butterworth, London, UK.

Malavé-López, J., Barbosa-Cánovas, G.V., and Peleg, M. 1985. Comparison of the compaction characteristics of selected food powders by vibration, tapping and mechanical compression. *J. Food Sci.* **50**, 1473–1476.

Mao, R., Tang, J., and Swanson, B.G. 2000. Texture properties of high and low acyl mixed gellan gels. *Carbohydr. Polym.* **41**, 331–338.

Mohsenin, N.N. 1986. "Physical Properties of Plant and Animal Materials". Gordonand Breach Science Publishers Amsterdam, The Netherlands.

Molina, M., Nussinovitch, A., Normand, M.D., and Peleg, M. 1990. Selected physical characteristics of ground roasted coffees. *J. Food Process. Preserv.* **14**, 325–333.

Moreyra, R. and Peleg, M. 1980. Compressive deformation patterns of selected food powders. *J. Food Sci.* **45**, 864–868.

Moreyra, R. and Peleg, M. 1981. Effect of equilibrium water activity on the bulk properties of selected food powders. *J. Food Sci.* **46**, 1918–1922.

Mort, P.R., Sabia, R., Niesz, D.E., and Riman, R.E. 1994. Automated generation and analysis of powder compaction diagram. *Powder Technol.* **79**, 111–119.

Murthy, C.T. and Bhattacharya, S. 1998. Moisture dependent physical and uniaxial compression properties of black pepper. *J. Food Eng.* **37**, 193–205.

Niediek, E.A. 1971. Studies on crushing of crystal sugar. *Z Zuckerindus.* **21**, 432–439.

Niranjan, K. 1995. An appraisal of the characteristics of food mixing. *In* "Food Process Design and Evaluation" (R.K. Singh, ed.). Technomics, Lancaster, PA, USA.

Niranjan, K., de Alwis, A.A. (1993). Agitation and agitator design. *In* "Encyclopedia of Food Science, Food Technology and Nutrition" (R. Macrae, R.K. Robinson, and M. Sadler, eds), Vol. 1. Academic Press, London.

Normand, M.D. and Peleg, M. 1986. Determination of the fractal dimension of a particle silhouette using image-processing techniques. *Powder Technol.* **45**, 271–275.

Nuebel, C. and Peleg, M. 1994. A research note: Compressive stress-strain relationships of agglomerated instant coffee. *J. Food Process Eng.* **17**, 383–400.

Nyström, C. and Karehill, P.G. 1996. The importance of intermolecular bonding forces and the concept of bonding surface area. *In* "Pharmaceutical Powder Compaction Technology" (G. Alderborn and G. Nyström, eds), p. 17. Marcel Dekker, New York.

Ollett, A.L., Kirby, A.R., Parker, R., and Smith, A.C. 1992. A comparative study of the effects of water content on the compaction behavior of some food materials. *Powder Technol.* **75**, 59–65.

Onwulata, C.I., Smith, P.W., and Holsinger, V.H. 1995. Flow and compaction of spray-dried powders of anhydrous butteroil and high melting milkfat encapsulated dissacharides. *J. Food Sci.* **60**, 836–840.

Onwulata, C.I., Smith, P.W., and Holsinger, V.H. 1998. Properties of single- and double-encapsulated butteroil powders. *J. Food Sci.* **63**, 100–103.

Panelli, R. and Filho, F.A. 2001. A study of a phenomenological compacting equation. *Powder Technol.* **114**, 255–261.

Parikh, D.P. (ed.) 1997. "Handbook of Pharmaceutical Granulation Technology", no. 81, "Drugs and Pharmaceutical Sciences". Marcel Dekker, New York.

Paronen, P. and Ilkka, J. 1996. Porosity: Pressure functions. *In* "Pharmaceutical Powder Compaction Technology" (G. Alderborn and G. Nyström, eds), p. 55. Marcel Dekker, New York.

Peleg, M. 1978. Flowability of food powders and methods for its evaluation. A review. *J. Food Process. Eng.* **1**, 303–328.

Peleg, M. 1983. Physical characteristics of powders. *In* "Physical Properties of Foods" (M. Peleg and E.B. Bagley, eds), pp. 293–324. Van Nostrand Reinhold/ AVI, New York.

Peleg, M. 2004. Physical Properties of Food Powders. Encyclopedia Of Life Support Sciences (EOLSS). Topic 5.10.1.5. Eolss Publishers Co Ltd, Oxford, U.K. In Press.

Peleg, M. and Hollenbach, A. 1984. Flow conditioners and anticaking agents. *Food Technol.* **38**, 93–102.

Peleg, M. and Mannheim, H. 1973. Effect of conditioners on the flow properties of powdered sucrose. *Powder Technol.* **7**, 45–50.

Peleg, M., Mannheim, H., and Passsy, N. 1973. Flow properties of some food powders. *J. Food Sci.* **38**, 959–964.

Peleg, M. and Normand, M.D. 1985a. A research note: Characterization of the ruggedness of instant coffee particle shape by natural fractals. *J. Food Sci.* **51**, 829–831.

Peleg, M. and Normand, M.D. 1985b. Mechanical stability as the limit to the fractal dimension of solid particle silhouettes. *Powder Technol.* **43**, 187–188.

Pietsch, W. 1991. "Size Enlargement by Agglomeration". John Wiley & Sons, Chichester, UK.

Pietsch, W. 1994. "Parameters to be considered during the selection, design, and operation of agglomeration systems. Preprints of 1st International Particle Technology Forum. Part I". AIChE, New York.

Pietsch, W. 1999. Readily engineer agglomerates with special properties from micro- and nanosized particles. *Chem. Eng. Prog.* **95**, 67–81.

Ramberger, R. and Burger, A. 1985. On the application of the Heckel and Kawakita equations on powder compaction. *Powder Technol.* **43**, 1–9.

Rennie, P.R., Chen, X.D., Hargreaves, C., and Mackereth, A.R. 1999. A study of the cohesion of dairy powders. *J. Food Eng.* **39**, 277–284.

Risch, S.J. 1995. Review of patents for encapsulation and controlled release of food ingredients. *In* "Encapsulation and Controlled Release of Food Ingredients" (S.J. Risch and G.A. Reineccius, eds), pp. 197–203. American Chemical Society, Washington, DC.

Roberts, A.W. 1998. Bulk solid and conveyor belt interactions for efficient transportation without spillage. *Bulks Solids Handling* **18**, 49–57.

Roberts, A.W. 2001. Recent developments in belt conveying—bulk solid and conveyor belt interactions. *In* "Handbook of Conveying and Handling of Particulate Solids" (A. Levy and H. Khalman, eds), pp. 225–233. Elsevier, New York.

Rumpf, H. 1962. The strength of granules and agglomerates. *In* "Agglomeration" (W.A. Knepper, ed.), pp. 379–418. Interscience Publishers, New York.

Sandor, B.I. 1998. Mechanics of materials. *In* "The CRC Handbook of Mechanical Engineering" (F. Kreith, ed.), pp. 1–67. CRC Press, Boca Raton, FL.

Scherge, M. and Gorb, S.N. 2001. "Biological Micro- and Nanotribology. Nature's Solutions". Springer-Verlag Berlin Heidelberg, Germany.

Schubert, H. 1981. Principles of agglomeration. *Int. Chem. Eng.* **6**, 1–32.

Schubert, H. 1987. Food particle technology. Part I: Properties of particles and particulate food systems. *J. Food Eng.* **6**, 22–26.

Shahidi, F. and Han, X.Q. 1993. Encapsulation of Food Ingredients. *Crit. Rev. Food Sci. Nutr.* **33**, 501–547.

Shinohara, K. 1997. Segregation of particles. *In* "Powder Technology Handbook" (K. Gotoh, H. Masuda, and K. Higashitani, eds), pp. 383–393. Marcel Dekker, New York.

Shipway, P.H. and Hutchings, I.M. 1993. Attrition of brittle spheres by fracture under compression impact loading. *Powder Technol.* **76**, 23–30.

Scanlon, M.G. and Lamb, J. 1995. Fracture mechanisms and particle shape formation during size reduction of a model material. *J. Material Sci.* **30**, 2577–2583.

Simons, S.J.R. 1996. Modeling of agglomerating systems: From spheres to fractals. *Powder Technol.* **87**, 29–41.

Sone, T. 1972. Consistency of foodstuffs. D. Reidel Pub. Co, Dordrecht, Holland.

Swyngedau, S., Nussinovitch, A., Roy, I., Peleg, M., and Huang, V. 1991. Comparison of four models for the compressibility of breads and plastic foams. *J. Food Sci.* **56**, 756–759.

Teunou, E., Fitzpatrick, J.J., and Synnot, E.C. 1999. Characterization of food powder flowability. *J. Food Eng.* **39**(1), 31–37.

Thomson, F.M. 1997. Storage and flow of particulate solids. *In* "Handbook of Powder Science and Technology" (M.E. Fayed and L. Otten, eds), pp. 389–486. Chapman and Hall, New York.

Van Heyst, T.L. 1983. "System design for the commercial compression of dried foods". M. S. Thesis. Department of Mechanical Engineering, University of Massachusetts, MA.

Williams, J.C. 1976. The segregation of particulate materials: A review. *Powder Technol.* **15**, 245–251.

Yan, H. and Barbosa-Cánovas, G.V. 1997. Compression characteristics of agglomerated food powders: Effect of agglomerated size and water activity. *Food Sci. Technol. Int.* **3**, 351–359.

Yan, H. and Barbosa-Cánovas, G.V. 2000. Compression characteristics of selected food powders: The effect of particle size, mixture composition, and compression cell geometry. *In* ''Selected physical properties of food powders'', Doctoral Thesis, pp. 82–113. Biological Systems Engineering Department, Washington State University.

Yan, H. and Barbosa-Cánovas, G.V. 2001a. Attrition evaluation for selected agglomerated food powders: The effect of agglomerate size and water activity. *J. Food Process Eng.* **24**, 37–49.

Yan, H. and Barbosa-Cánovas, G.V. 2001b. The effect of padding foam on the compression characteristics of some agglomerated food powders. *Food Sci Technol. Int.* **7**, 417–423.

Yan, H., Barbosa-Cánovas, G.V., and Swanson, B.G. 2001. Density changes in selected agglomerated food powders due to high hydrostatic pressure. *Lebensm.-Wiss. U. Technol.* **34**, 495–501.