FROM BATCH TO PRESSED TILE: MECHANICS AND SYSTEM MICROSTRUCTURAL CHANGES

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Fired tile quality depends very strongly on the characteristics developed in the as-pressed tile. The achievement of ever-higher-quality tile microstructures and customized properties can benefit from scientific analysis, models, and insights. This lecture is an overview of materials parameters and processes influencing the pressed green body: materials characteristics, developing a controlled slurry with essential additives; spray drying to form controlled agglomerates (granules); the relationship of agglomerate structure to slurry dispersion and rheology; agglomerate flow and packing in the die; agglomerate deformation and microstructure development when the tile is pressed. General scientific models and parameters established for pressed ceramics are described.

INTRODUCTION

Ceramic tile is commonly formed from a body of materials containing clay, flux, and refractory filler materials. Categories of tile products are indicated in Table 1. A broad range of products varying in dimensions, dimensional precision, strength, apparent porosity, surface texture, decorative coatings, and overall quality are produced by the tile industry. The manufactured tile product has a high ratio of surface area to thickness and the manufacturing process must be capable of producing this shape in a highly productive manner.

Table 1. Categories of Tile Products [1]
Porcelain impervious tile
Stoneware vitrified floor tile
Porous wall tile
Unglazed rustic stoneware floor tile

The tile market continues to be a growing market and especially so for higher quality products. Support continues for improved quality and manufacturing productivity. Even though manufacturing has undergone considerable changes in recent years, additional technological improvements will be needed.

Modern tile production requires predictive performance. Variation of dimensional stability among products or within the same product type is one defect that currently most affects ceramic tile quality [2]. Another is the variation within tile characteristics such as pore size and apparent porosity, and properties such as strength and chipping resistance. Much experience shows a high correlation between variation within a production lot of unfired tile and variation within the fired tile. For reasons of uniformity across a range of product characteristics and properties, coupled with high product yield and productivity, uniaxial dry pressing is now the most widely used forming method.

[1] SANCHEZ, E.; GARCIA, J.; SANZ, V.; AND OCHANDIO, E. *Raw Material Selection Criteria for the Production of Floor and Wall Tiles.* Tile Brick Int. 6(4) 15-21, 1990.

[2] NEGRE, F., AND SANCHEZ, E. *Advances in Spray-Dried Powder Processing for Tile Manufacture.* pp. 169-181 in Science of Whitewares. Edited by V. Henkes, G Onoda, and W. Carty. The American Ceramic Society, Westerville, OH USA. 1996.

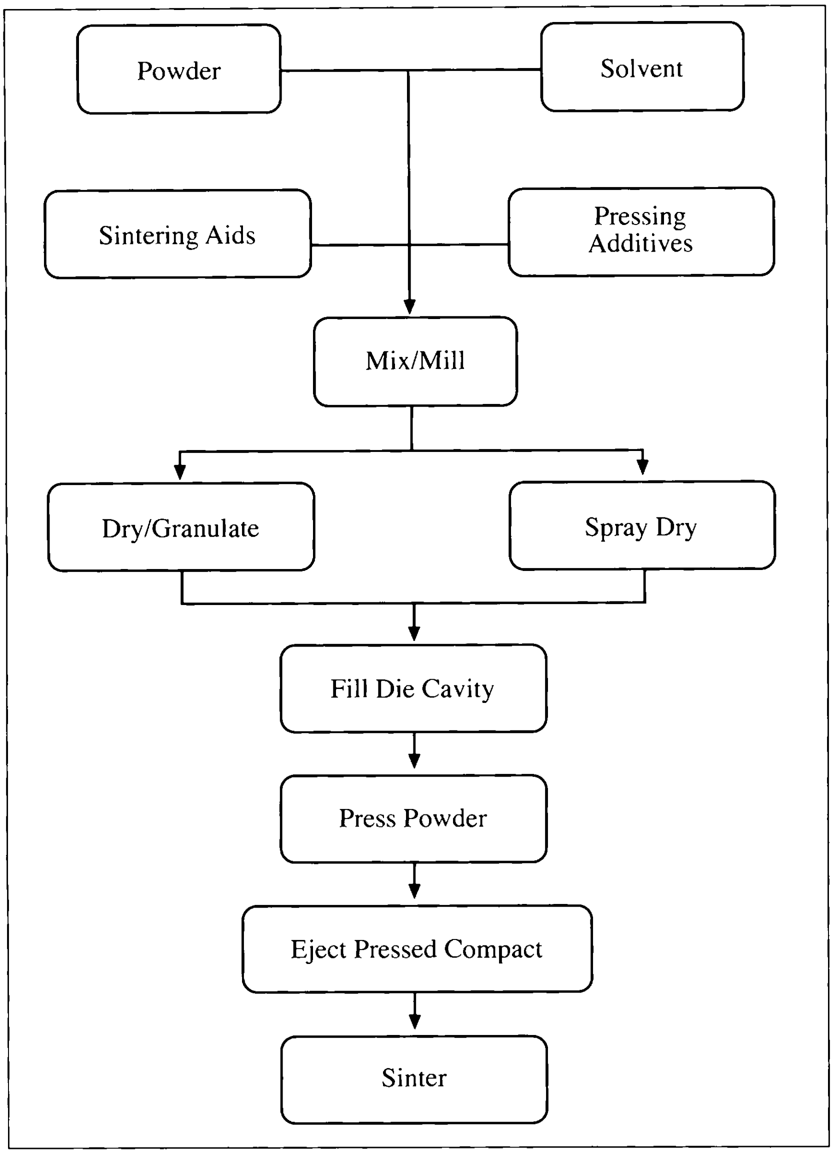

Figure 1. *General processing flow diagram for forming by powder pressing.*

A general processing flow diagram for forming a ceramic product using uniaxial dry pressing is shown in figure 1. The batch is comprised of powders and granular materials that on firing consolidate and shrink into a more dense mass. For ceramic tile, the powder is dispersed clay and ground minerals. Water is the solvent for reasons of availability, cost and safety. Although called dry pressing, the batch commonly contains moisture in the range of 4 to 8 wt%. Pressing additives would be those introduced specifically to assist in developing the required pressed density and microstructure with a minimum of variability over the piece and within a production lot of tile. These additives may be organic or inorganic in nature, and may be used in combination. Mixing of the batch components is a very critical step that strongly influences product homogeneity. Dispersion of agglomerated batch ingredients is a critical component of the mixing operation. Milling, which may be combined with mixing in one operation, is used to reduce the sizes of larger primary particles and aggregates, to reduce the average particle size of the batch, and to chemically alter surfaces of particles on a microscopic scale. A granulated free-flowing material is needed for rapid dry pressing. Granulation is accomplished using either a "dry" process or in a wet process that is commonly spray drying. The controlled agglomerates produced are called granules. Granules are mechanically pressed to form the unfired tile. During the pressing operation, granules must deform in a controlled manner enabling the formation of a more dense, shaped product with sufficient strength for subsequent handling including any surface finishing operations.

BATCH MATERIALS

The batch of materials must be capable of being processed efficiently into a feed material that can be conveniently pressed and then fired to produce a tile product with controlled density, strength and surface characteristics. It is very desirable that the pressed density not be highly sensitive to small variations in prior processing operations. Also, the tile should have a wide firing range so that the final dimensions are relatively insensitive to small differences in temperature during firing.

A batch for ceramic tile will commonly contain a ground flux such as feldspar, one or more clay materials, and ground calcined material. Materials used range widely depending on the product type and nature of the chemical impurities in the raw materials. The flux is a filler material in the green body and later provides vitrification for sintering at a moderate temperature on firing. Clay facilitates particle sliding and cohesive strength when pressed and modifies the pyroplastic behavior during firing. Refractory materials serve as fillers. Kaolin clays contain impurity particles such as hematite and titania and feldspars contain quartz and traces of iron oxide and magnesia. The other batch components may be refractory minerals such as a silica sand, calcined clay materials (chamotte), natural and calcined ceramic colorants, accessory fluxes, etc . Calcium carbonate is sometimes added in wall tile bodies. Impurity phases associated with raw materials may be free particles or an impurity contained within larger particles.

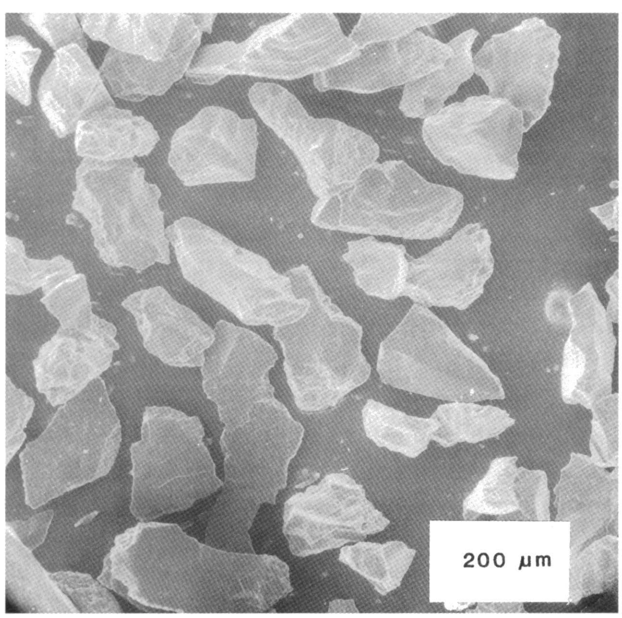

Figure 2. *Rough, angular particles of nonplastics in batch.*

Batch materials produced by crushing and grinding are commonly of an angular shape and have relatively rough surfaces when viewed on a microscopic scale, as is shown in figure 2. These materials act as a filler in the green body and are commonly called the "non-plastic" component. Clay materials when dispersed commonly contain a significant fraction of particles in the finer fractions that are plate-like in shape and have very smooth faces. The clay is commonly called the "plastic" component but large clay mineral aggregates act as a non-plastic filler. The coating of finer plate-like clay on larger particles with rough surfaces reduces friction and provides for easier particle sliding during pressing. A matrix of fine clay coating larger particles provides cohesive strength in the pressed tile. The clay must be selected both for green body processing requirements and for firing requirements. Specifications of starting materials include the chemical analysis, loss on ignition, x-ray analysis, particle size distribution, specific surface area, particle density, and in addition for clays the cation exchange capacity, suspension pH, and dry modulus of rupture.

BATCH PROCESSING

The batch of materials must be processed to develop feed material with an appropriate behavior for pressing. Processing goals for ceramic tile are listed in Table 2.

Table 2. Processing Goals for Ceramic Tile [2]

Disperse and distribute clay materials in the batch. Wet processing enables chemical in addition to physical forces to assist dispersion and alter mechanics in the machinery.
Reduce larger particle sizes enabling liberation of impurity particles. Wet processing is more complete.
Improve homogeneity. Wet processing commonly improves mixedness as a consequence of improved dispersion and greater mobility of particles for more complete intermingling. This is especially important for minor additives/components.
Increase specific surface area of materials via reduction of particle sizes. This increases batch reactivity during mixing and on firing.
Uniformity of granule character and pressability batch to batch. Low variation of composition including moisture within granulated material.
Produce granules of appropriate density and shape with high yield within the size range 500-125 μm.

Dry process

In the "dry process" the batch is prepared with a moisture content approximating that needed for pressing. Advantages of the dry process are a smaller capital investment and lower running and production costs. Unit operations are dispersion, mixing, and granulation.

[2] NEGRE, F., AND SANCHEZ, E. *Advances in Spray-Dried Powder Processing for Tile Manufacture.* pp. 169-181 in Science of Whitewares. Edited by V. Henkes, G Onoda, and W. Carty. The American Ceramic Society, Westerville, OH USA. 1996.

Dispersion reduces the sizes of agglomerates of the raw materials and mixing combines and intermingles the particles. It is important to note that dispersion and mixing reduce the scale of segregation (SOS) of components within the batch, which is a prerequisite for reducing the scale of inhomogeneity (SOI) within the microstructure of the tile. The scale of inhomogeneity is larger than the scale of segregation, ie.

$$SOI > SOS \qquad (1)$$

Undispersed material or incompletely distributed material will have a relatively large SOS and create a region in the tile of large SOI.

A particular level of reproducible particle dispersion throughout the batch is a processing requirement. Particle dispersion is generated by physical forces and in wet processing is facilitated significantly by chemical forces. Particle size reduction (comminution) commonly precedes mixing when using the dry process. The characteristics of these "preprocessed" materials such as particle size distribution must be very well controlled. A very high-energy mixing device is needed to efficiently disperse and uniformly intermingle the fine particles with larger ones. Attention to the moisture content within feed powders that strongly influences agglomerate strength is required. The dispersion and premixing of a minor additive of high specific surface area, such as a ceramic colorant, within another fine batch component, before mixing all materials, may be required to achieve its homogeneous distribution.

Material selection, considering particle dispersability is extremely important when using a dry process to produce higher quality products. A batch particle size distribution of the AFDZ type described below is needed to achieve a high density in the pressed tile. The granulating step must also be carefully controlled. Using the dry process, a very significant concern is the capability of the processing machinery to introduce moisture uniformly throughout the volume of the batch and produce granules of controlled size, shape, and density (Figure 3).

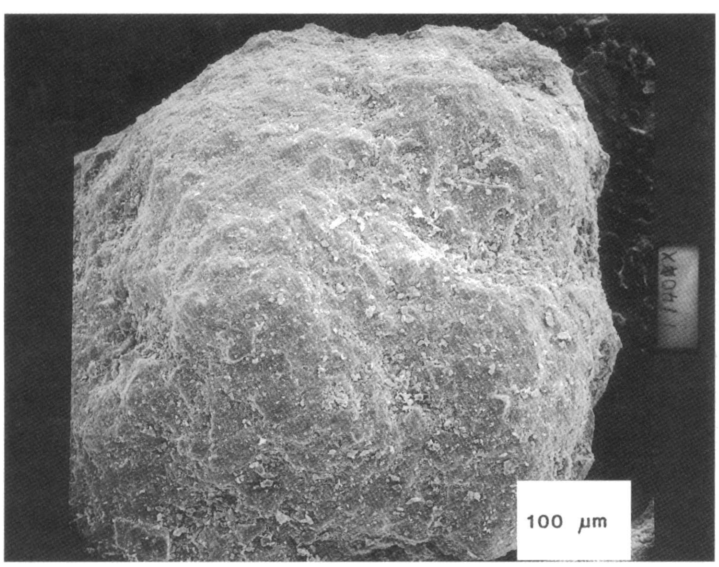

Figure 3. *Scanning electron micrograph of a granule made in a "dry process."*

It is reported that presently this route can not meet product quality and performance requirements for the more-upscale products[2]. Upgrades in the dry process seem possible. Here improvements in machinery design and performance and correlation of processing parameters to granulate performance are needed.

Wet process

In the "wet process" the batch is first processed in slurry form. Milling and mixing can be accomplished in one step such as by wet ball milling. Tile bodies processed as slurries are mixed, dispersed, pumped, sieved, and then spray dried. Processing costs per unit of granulated product are reduced significantly when the slurry has a high solids loading and relatively less water must be eliminated. Near-surface ions on particles may be dissolved into adsorbed water films. Particle surfaces may become hydrated and adsorb ions and additives, becoming electrically charged, and also adsorb colloidal (submicron) substances. A particular, controlled amount of submicron material is needed to produce adequate slurry properties. A general goal is to increase the solids content as much as possible while maintaining adequate slurry flow properties. Achieving this goal requires close control of chemical deflocculation and the particle size distribution in the processed slurry.

Particle size distribution. The water requirement for slurry processing depends on both the range of particle sizes and the form of the particle size distribution. Particle sizes in tile bodies are commonly finer than 50-100 μm and range down to about 0.1 μm. A distribution of particle sizes that packs more efficiently has a lower water requirement for flow because relatively less of the water is present in interstices where the water does not facilitate flow.

The close-packing size distribution for the tile body is approximated by the AFDZ equation [3,4]

$$\text{Mass Fraction Finer} = \frac{D^n - (D_{min})^n}{(D_{max})^n - (D_{min})^n} \qquad (2)$$

Where D is any particle size and D_{max} and D_{min} are the maximum and minimum particle sizes, respectively. An equivalent equation is

$$\log[\text{Mass Fraction Finer}] = \log[D^n-(D_{min})^n] - \log[(D_{max})^n-(D_{min})^n] \qquad (3)$$

By plotting the cumulative size distribution vs. particle size on log-log scale, the approximation of the particle size distribution of the tile body to this distribution function is easily seen (see Figure 4). For the AFDZ distribution, the larger 80% of the size distribution should appear linear on the graph. The measured slope of the linear region is the distribution modulus "n". Funk and Dinger [4] have shown that the maximum packing density occurs when n is 0.35 - 0.40.

[3] Ortega, F. S.; Pileggi, R. G. ; Sepulveda,P.; and Pandolfelli, V. C. *Optimizing Particle Packing in Powder Consolidation.* Am. Ceram. Soc. Bull. 78(8) 106-111, 1999.

[4] Lopez-Begue,L. A.; Dinger, D. R.; and Funk, J. E. *PSD Affects Rheology of Triaxial Porcelains.* Am. Ceram. Soc. Bull. 76(9) 83-87, 1997.

The blending of appropriate nonplastics of larger particle sizes with clays of finer sizes may produce a batch size distribution of the AFDZ type that can pack to a relative density of 80% (2.0 g/cm[3]) in the pressed tile. The wet high-energy dispersion of clays and grinding and dispersion of all raw materials by ball milling, increases the proportion of submicron particles that can significantly improve the particle packing.

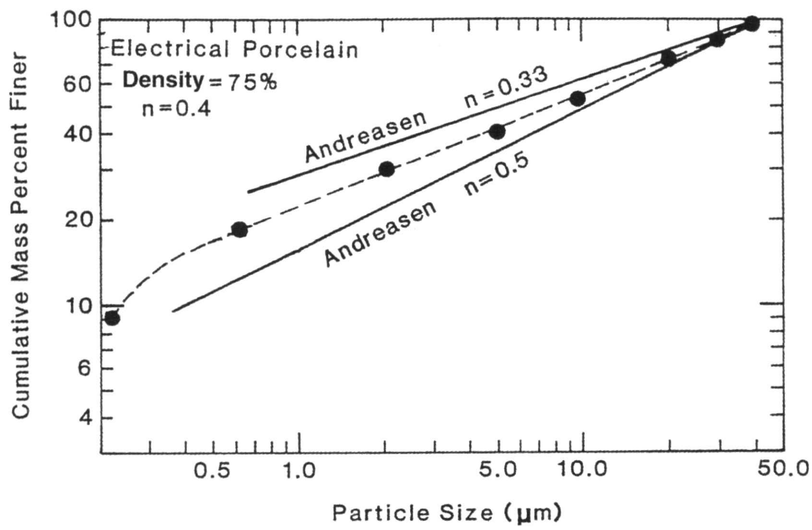

Figure 4. *Graph for a porcelain body with a particle size distribution approximating the AFDZ type.*

Table 3. Isoelectric Points of Materials Used in Tile Bodies [5]

Material	Nominal Composition	IEP
Quartz	SiO_2	2
Albite	$Na_2O.Al_2O_3.6SiO_2$	2
Talc	$4SiO_2.3MgO.H_2O$	2-3
Orthoclase	$K_2O.Al_2O_3.6SiO_2$	3-5
Kaolin	$Al_2O_3.SiO_2.2H_2O$	4-7
Mullite	$3Al_2O_3.2SiO_2$	7-8
Hematite	Fe_2O_3	6-9
Alumina	Al_2O_3	7-10
Barium Carbonate	$BaCO_3$	10-11

The dense packing distribution is important not only to minimize the water demand of the slurry and increase the tile's pressed density and strength, but also to provide greater resistance to pyroplastic deformation when the tile is fired.

Chemical deflocculation. Numerous papers have been published on the deflocculation of slurries of ceramic tile bodies, and general principles are now well established. However, the chemical behavior of the raw materials for tile may vary widely and some empirical experimentation is needed to develop a slurry with appropriate rheological properties, and to stabilize the slurry.

[5] Reed, J. S.: *Principles of Ceramics Processing.* 2nd Edition. J. Wiley & Sons, Chichester, UK, 1995.

In an aqueous slurry, particle charging is the primary mechanism for deflocculation. Particles with like electrical charge repel and those with opposite charge attract. The charge on an oxide particle in water will vary with the pH of the aqueous solution in the slurry. The isoelectric point (IEP) of the surface is the pH at which the zeta potential near the surface (at shear plane for moving particles) is neutral. At a pH below the IEP, the particle is positively charged and at a pH above the IEP the particle will be negative (see Figure 5). IEP values for materials used in tile bodies are listed in Table 3, Impurities in a material and the dissolution of ions from particle surfaces can cause a shift in the IEP of a material.

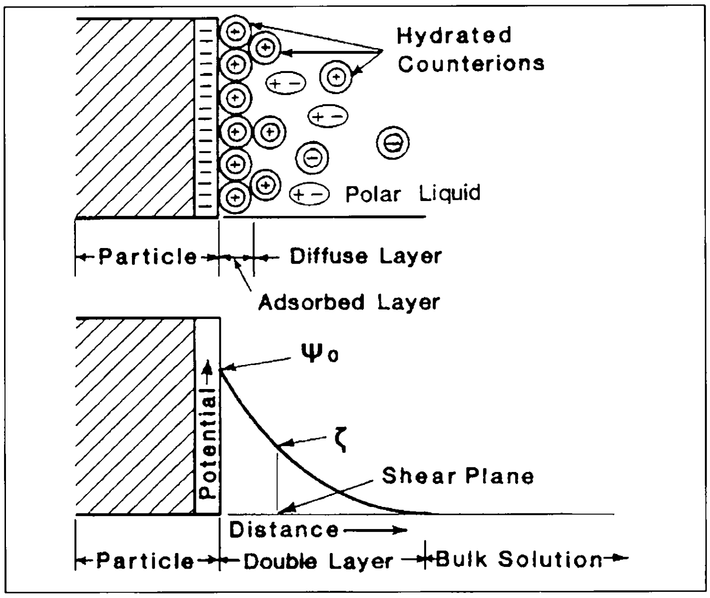

Figure 5. Ion distribution in liquid near a negatively charged surface.

Dissolution of ions from the surface of a particle may also cause a particle to become charged and may cause a shift in the IEP. An example is the dissolution of an alkali from a feldspar. Kaolinite is a special mineral with a plate-like shape; the faces and edges of the particle have a very different surface chemistry. Cations are adsorbed between faces in clay aggregates. On dispersing the kaolinite in water, desorption of charge-compensating cations causes the faces to be negatively charged over the range of pH common in slurries for tile processing (Figure 6). The edges of the kaolinite react quite differently to the addition of an acid or base.

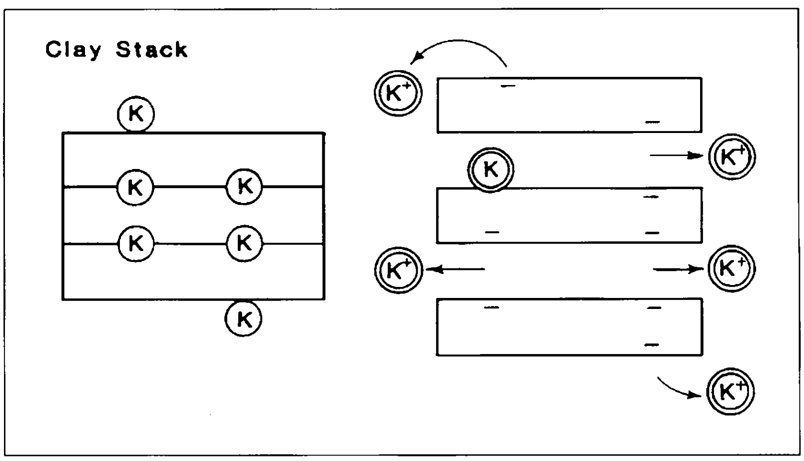

Figure 6. Charging of faces of kaolin particles on desorption of cations.

The addition of an acid/base to the slurry to shift the pH < or > IEP may cause the particles to become charged. Adsorption of negatively charged polymer ions such as an acrylate or phosphate (see Figure 7) on particles that have a slightly positive or neutral surface will produce a negative surface layer. Control of the solution pH is needed to maximize the polymer adsorption. Because of their higher specific surface area, submicron materials, and especially clays, will have a relatively higher influence for an equivalent weight fraction, and the IEP of the submicron material must especially be considered. A sample of the aqueous solution for pH measurement is easily obtained by decanting after centrifuging a small sample of the slurry.

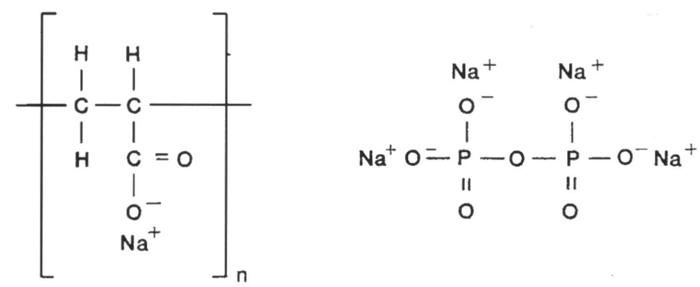

Figure 7. *Molecular structure of sodium polyacrylate and sodium pyrophosphate.*

Polymer anions such as acrylate, methacrylate, and silicate with a loop conformation produce a physical hindrance to the close approach of particles and are called electro-steric deflocculants. These polymer deflocculants can be purchased in grades that vary in molecular weight. Sodium phosphate and sodium silicate can be purchased in grades differing in alkali content. As seen in Table 4, solutions of the silicates have a higher pH in the range 10-12, whereas for solutions of sodium phosphate the pH decreases from about 11 to 6 with increasing phosphate content. These differences must be recognized when adjusting the slurry pH in order to maximize adsorption of the deflocculant on the particles. The crystalline form of the triphosphate may also influence dissolution and alter the deflocculating behavior [7].

Table 4. Characteristics of Phosphates and Silicates [6]

Phosphates	pyro	tripoly	hexameta
mol ratio P_2O_5/Na_2O	0.5	0.6	1.0
pH of 1% soln.	10.7	9.7	6.1
Silicates	meta	di	tri
mol ratio SiO_2/Na_2O	1.0	2.1	3.1
pH of 1% soln.	12.3	11.5	10.6

Cations in the water used for slurry preparation and dissolution of ions such as K^+, Mg^{2+}, Ca^{2+}, and Fe^{3+} from the surfaces of primary particles and soluble impurities may

[6] MANFREDINI, T.; PELLACANI, G. C.; POZZI,P.; BLASCO FUENTES, A.; AND NEGRE MEDALL, F. *Some General Consideration of the Rheological Behaviour of Aqueous Clay Suspensions: Dependence on the Physicio-Chemical properties of Inorganic Salts, Calcium (II) Ion Presence and Grinding Times.* Ind. Ceram. 9(2) 58-62, 1989.

[7] ANDREOLA, F.; POZZI, P.; AND RODRIGUES NETO, J.B. *Rheological Behavior of an STP Deflocculated Kaolin.* Am. Ceram. Soc. Bull. 77(12) 68-71, 1998.

complicate the deflocculation. More intense dispersion of the slurry in a high-energy mixer or ball mill will speed dissolution. Highly charged cations can adsorb on the surfaces of particles and reduce the negative surface charge and forces of repulsion. An effective deflocculant is one whose action predominates over the effects of impurity ions. The addition of a multivalent additive such as sodium polyacrylate, sodium polymethacrylate, or sodium phosphate is commonly required in industrial processing for at least two reasons. At the processing pH of the slurry, the acrylate molecule will be significantly dissociated with a large negatively charged group and free alkali ions. A multi-charged negative phosphate molecule may react with and precipitate cations such as Ca^{2+}. An acrylate or methacrylate may chelate the highly charged cation (sometimes called a sequestering action). Both mechanisms eliminate highly charged cations from interfering with deflocculation. Sodium silicate is thought to provide a protective colloidal silica gel assisting deflocculation, through the reaction

$$Na_2O \bullet nSiO_2 + H_2O = nSiO_2 + 2Na^+ + 2OH^- \qquad (4)$$

Grades with SiO_2/Na_2O in the range 1-2 are commonly used.

The combination of the electrosteric action and the sequestering of highly charged cations can commonly provide powerful deflocculation when using commercial materials and industrial processing water. The particular chemical and physical actions of a phosphate, silicate, acrylate, or methacrylate on materials in a multi-component slurry may vary somewhat. Each additive can be purchased in grades having a different molecular weight (chain length) and alkali content. Although added in an amount of 0.3-0.5 wt% of the inorganic materials in the batch, the cost of the additives is significant. For reasons of deflocculating effectiveness, slurry stability over time, and minimal cost, two deflocculants in combination such as a tripolyphosphate and metasilicate mixture may provide satisfactory deflocculation at a lower cost.

SPRAY DRYING

Granules of controlled density, shape, size range, and moisture content are required for rapid and uniform filling of the die and to produce pressed tile with a uniform microstructure and adequate strength for handling. Spray drying is now widely used for converting the slurry into controlled granulated feed for dry pressing. Spray drying is a three step process involving both droplet drying and granule formation. The process begins with atomization of the slurry feed into a spray of droplets. A heated gas stream suspends the droplets, evaporates water leaving behind agglomerates of solid particles, additives, and moisture having a size distribution similar to that of the droplets. Dried agglomerates separated from the drying air are collected as granulated feed material for pressing. Granules produced are more often hollow in shape as is shown in Figure 8.

Details of spray drying machinery and options are not presented here. For ceramic tile production where large quantities of material are needed, a mixed flow fountain-type dryer with multiple nozzles is commonly the practice. Control of the slurry solids content, density (slurry may have occluded air bubbles), flow properties, maintenance of nozzle geometry, the temperature and humidity of inlet air, and the flow conditions within the dryer are required to minimize variation in the granulated product over time. Slurry control parameters include the poured density, solids content, and flow properties. More important operational parameters for controlling the moisture content of the granules are maintaining a constant drying air temperature and a uniform slurry feed rate.

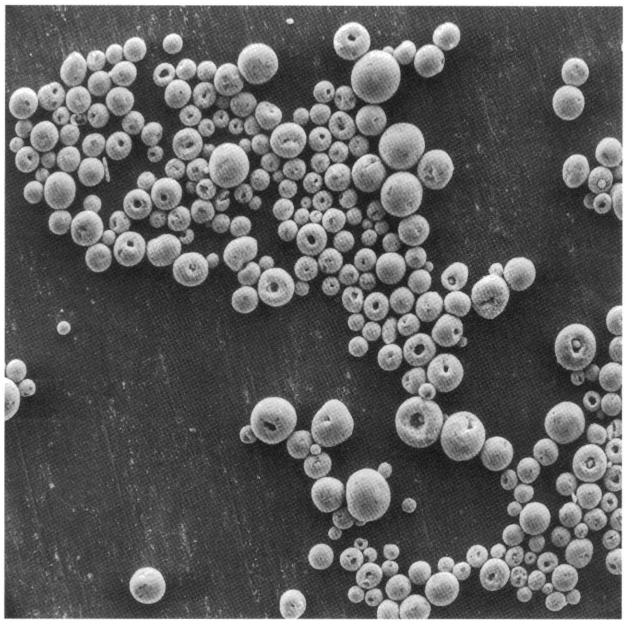

Figure 8. *Granules with deep cavity (hollow granules) formed by spray drying.*

Walker [8] recently established the dependence of granule characteristics on slurry flow properties measured up to the very high shear rates of 10^{-6} s^{-1} that occur during droplet formation. The mean granule size correlated directly with the viscosity measured at the high shear rate. Granule density (primary particle packing density in dried droplet) correlated inversely with slurry yield stress measured at zero shear rate. The yield stress is controlled by the solids content and the concentration of deflocculant.

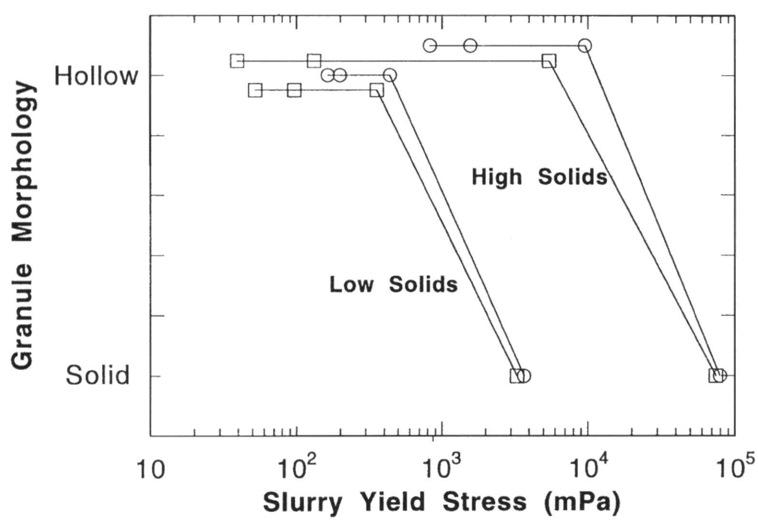

Figure 9. *Shape of spray-dried granules depends on solids content and yield stress of slurry.*

Granule shape also depended directly on the properties of the slurry. Granules with a large cavity were produced on spray drying slurries at maximum deflocculation having a low yield stress, as is shown in Figure 9. "Solid" granules containing only uniform small pores throughout were produced on spray drying a slurry containing less deflocculant that had a higher yield stress. These observations are explained by the model for granule

[8] WALKER, W.J.; *PhD Thesis,* NY College of Ceramics, Alfred University, Alfred, NY USA.

formation shown in Figure 10. Small flocculation forces between particles in the droplet restrict particle mobility and can enable relatively rapid formation of a network of connected particles that withstands the capillary forces causing cavity formation. Particle packing in the solid granules was observed to be slightly lower as would be expected for the lower deflocculant concentration. But for the different granules, the bulk fill densities after pouring granules into a graduated cylinder were nearly identical because granules with a higher packing density of primary particles were hollow.

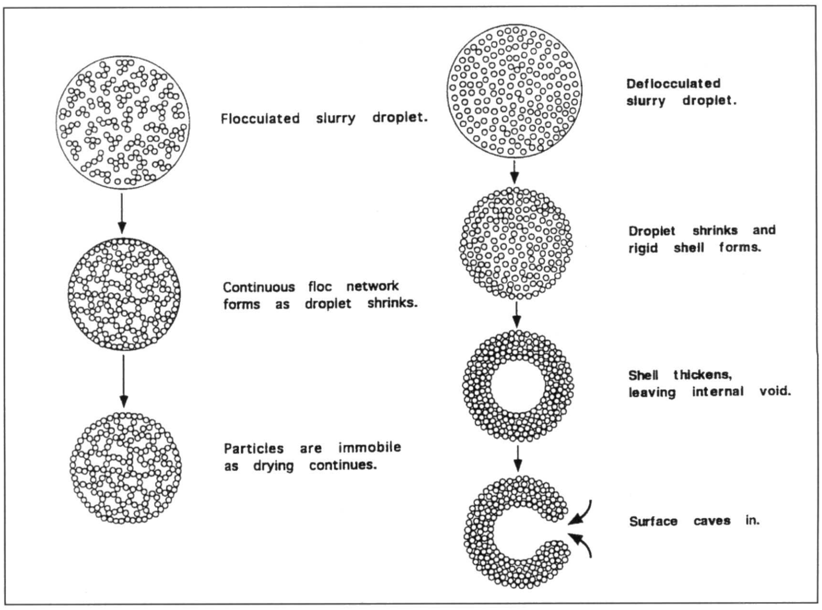

Figure 10. *Particle model for the formation of "solid" and "hollow" granules when droplets dry.*

FEEDING THE PRESS

Die filling is the most important pressing step [9]. Uniform flow of the granulated feed material is absolutely essential for the production of pressed tile of uniform pressed density. Flowability depends directly on the characteristics of the granulated feed indicated in Table 5, and strongly influences die filling behavior.

Table 5. Parameters Influencing Mass Flow of Granules.

Characteristic	Desired feature
granule shape	spherical
granule density	high
average granule size	large
size range of granules	fines < 74 μm removed; controlled
size distribution of granules	close packing type
granule surface	smooth
intergranule forces	no adhesion

Van der Waals forces and adhesion produced by surface moisture may cause granules to adhere. Offsetting this is the weight of the granule that can motivate its flow.

[9] AMOROS, J.L.; BLASCO, A.; ENRIQUE, J.E.; AND NEGRE, F.: *Caracteristicas de polvos cerámicos para prensado.* Bol. Soc. Esp. Ceram. Vidr., Vidr., 26(1) 31-37. 1987.

Dynamic flow involves the rolling and tumbling of many granules. Granules that are large, spherical, dense, with smooth non-adhering surfaces will have better flowability. When considering mass/time for flow, a granule size distribution with a moderately broad size range larger than about 74-125 μm that packs relatively efficiently is desired. Also, the moisture content must be controlled within narrow limits.

A high density of the granulated material in the die (fill density) is desired for pressing. It is generally observed that a higher fill density correlates with a higher mass flow rate and smaller angle of repose. Whereas a small percent of fines < 74 μm might be expected to increase the packing efficiency, attractive forces retard the flow of small granules of low mass and reduce the fill density and flow rate, even when present in only a few percent. These fine granules must be removed by screening. The pore size distribution in the filled die will be bimodal with smaller pores within granules and larger pores between granules. When granules themselves contain a relatively large pore, the pore size distribution will be trimodal. It is important to note that a higher fill density indicates more efficient packing and a smaller average interstitial pore size.

Feeding the granules into the die must be controlled well to obtain tile having a uniform pressed density. Pressing should begin with a uniformly filled die cavity. Uniforn filling depends on the geometry of the feeding hardware and granule flowability. Control of variation in the granule characteristics indicated in Table 5 is required to maintain flowability. A common problem is a lower fill density near edges and in corners of the die. This problem is reduced by controlling the speed of the filling shoe and the spacing and angle of baffles directing granule flow into the die cavity. Uniformity of granulate fill in the die is more of a problem when pressing larger-size tile. The importance of achieving a uniform pore size throughout the granulated fill including edges and corners can not be overemphasized.

PRESSING

The pressing process must also be closely controlled. A high pressed density is essential for reasons of higher off-the-press strength and a lower firing shrinkage. Density uniformity is required to minimize shape distortion (wedging) on firing. A reproducible pressed density and low firing shrinkage are required to maintain a low variation in fired dimensions, and especially so for larger tile sizes. The pressed product should be free of surface and edge defects and internal laminations. Defects in the pressed tile are incompletely eliminated during firing.

The increase in density on pressing a granulated material proceeds in three stages, as is indicated in Figure 11 [5]:

In Stage I, a small increase in the bulk density of the granulated fill in the die may occur from the slight rearrangement and sliding of granules with the first increment of applied pressure. This density increase of a few percent for free-flowing feed material is nearly zero when die filling is assisted using mechanical vibration.

Stage II begins when the pressure at contacts between granules is sufficient to cause particles within granules to slide, and the granules deform into neighboring interstices reducing the volume of the larger interstitial pores. The point of change

[5] Reed, J. S.: *Principles of Ceramics Processing.* 2nd Edition. J. Wiley & Sons, Chichester, UK, 1995.

of slope on the density vs. applied pressure diagram may be taken as an apparent yield pressure (Py) of the granules. Note that the density of the compacted granules at a particular pressure in Stage II is lower when the granules have a larger Py. As is indicated in Table 6, the apparent yield point varies inversely with the moisture content of the granules up to about 8 wt% in common tile compositions. Granulate feed of lower moisture content will be more resistant to deformation. Granules containing a relatively low content of clay will be less plastic and may have a larger minimum Py. The moisture content for peak density has been shown to decrease with a decreasing clay content [9]. In pressing tile, the pressing pressure is commonly less than 60 MPa. This pressure is not sufficient to produce an equivalent pore structure when the Py of granules varies significantly. Granulate feed that fills the die uniformly and deforms readily is needed to obtain pressed tile of uniform microstructure.

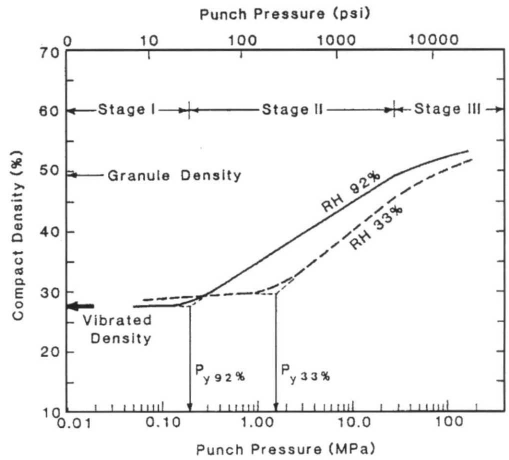

Figure 11. *General compaction diagram for powder granules having different yield points (Py).*

Table 6. Dependence of Apparent Yield Point of Granulated Feed (125-500 μm) on Moisture Content for a Typical Tile Body [2]

Moisture (wt%)	Py (MPa)
0.5	1.2
1	0.9
2	0.7
4	0.5
6	0.4
8	0.34

Stage III begins when the pore size in the compact becomes quite narrow and its density surpasses that of the feed granules. Elastic compression of granules begins in Stage II and increases rapidly in Stage III. Differential expansion (elastic springback) on ejecting the pressed tile may produce a crack or lamination when the

[2] NEGRE, F., AND SANCHEZ, E. *Advances in Spray-Dried Powder Processing for Tile Manufacture.* pp. 169-181 in Science of Whitewares. Edited by V. Henkes, G Onoda, and W. Carty. The American Ceramic Society, Westerville, OH USA. 1996.

[9] AMOROS, J.L.; BLASCO, A.; ENRIQUE, J.E.; AND NEGRE, F.: *Caracteristicas de polvos cerámicos para prensado.* Bol. Soc. Esp. Ceram. Vidr., Vidr., 26(1) 31-37. 1987.

local stress exceeds the strength in that region. Factors that cause a lower strength are discussed below. A larger apparent springback will produce a larger stress in the partially ejected tile. The springback will be higher for a higher pressing pressure and when using granules of lower moisture content. A body containing more of plate-like particles will generally exhibit more elastic springback and relatively more axial springback (anisotropic behavior). Bodies containing a high content of submicron clay are of relative low air permeability and when pressed rapidly the compressed air in pores can contribute a higher springback. Pressure gradients can also cause differential springback between regions in the tile.

Note in Figure12 that the fill density and pressed density for the tile body are about 40% higher than the values for the technical alumina body. These differences are a consequence of the close-packing AFDZ particle size distribution and wider range of particle sizes in the tile body. The tile body granules are of relatively lower porosity and the compacted granules are of much lower porosity. For the pressed density of 2.0 g/cm^3 the porosity is 23%. The high density indicates a relatively low linear shrinkage on firing.

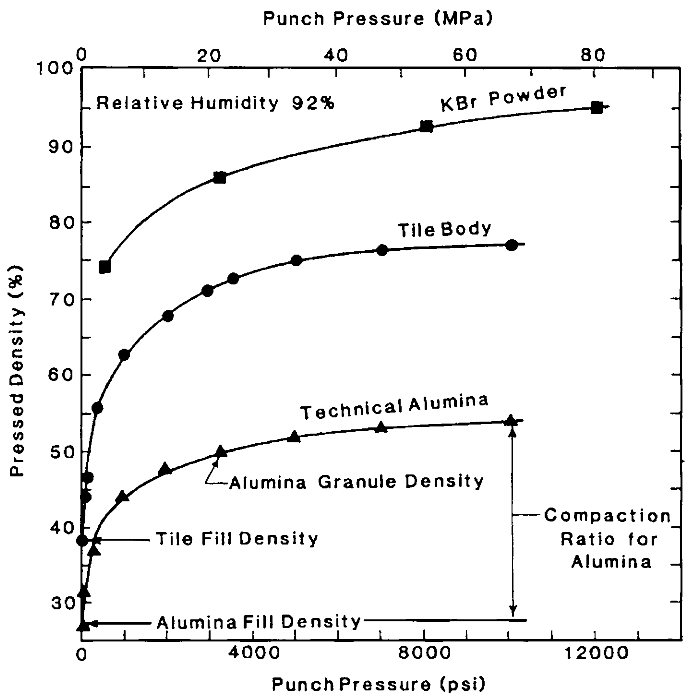

Figure 12. *Densification with pressure (linear scale) for tile and alumina pressing powders.*

Both bodies shown in Fig 12 press well and the compaction ratio is about 2.0 for each. The compaction ratio indicates the length of stroke and relative amount of compressed air that must be vented during pressing. Early in Stage II the air permeability depends on the size of interstices among granules that depends on the granule size and packing. Well into Stage II the air permeability depends on the size of the smaller pores in he compacted powder which are controlled particularly by the amount of fine clay particles. Air permeability is higher when using a relatively low clay content.

For uniaxial pressing near the end of Stage II, force is transmitted along pathways of contacts between larger granules, as is shown in Figure 13. This mechanism of pressure transmission aids in transmitting force into the depth of the compact, needed to produce a more uniform density.

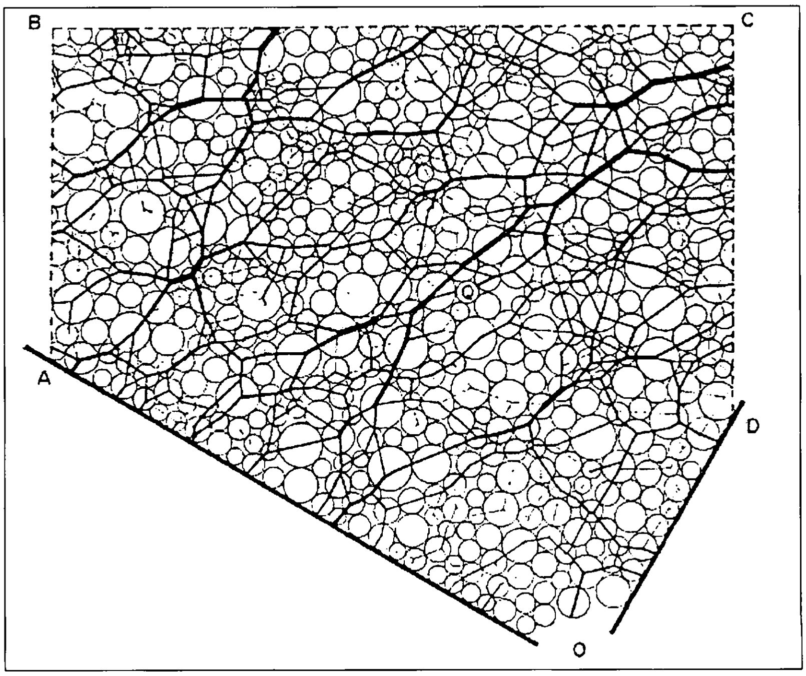

Figure 13. Pressure transmission modeling using computer simulation indicates some granules experience higher stress than others, indicated by line thickness. (from A. Drescher and G. de Josselin de Jong, J. Mech. Phys. Solids, 20 337-351, 1972.

PRESSED TILE STRENGTH

The mechanical strength of the pressed tile depends particularly on the bonding between clay particles and the absence of defects. A general relationship for the strength is [5, 10]

$$\text{Strength} = \frac{\text{function (number of interparticle contacts) (contact bond strength)}}{\text{function (defect size)}} \tag{5}$$

Bonding strength and number of interparticle contacts depend directly on the type and content of fine clay, the moisture content, and the pressed density. A higher pressing pressure commonly increases the density and the bonding of particles. The defect size limiting the strength in pressed tile corresponds to the size of flaws existing between large granules. Tile pressed using granules that do not deform easily during pressing will have a large defect size and a lower strength. When pressing well prepared granules, it is observed that the small granules deform and join together quite completely in the compact but large granules persist relatively undeformed. The stress at contacts between large granules is lower (see Figure 14) and the interstice where three large granules intersect remains as a relatively large pore defect in the compact.

The size of the large triangular interstices intersecting two or more weakly bonded large granules, remaining after pressing, is a function of the maximum granule size in the

[5] REED, J. S.: *Principles of Ceramics Processing.* 2nd Edition. J. Wiley & Sons, Chichester, UK, 1995.

[10] AMOROS, J.L.; FELIU, C.; GINES, G.; AND AGRAMUNT, J.V.: *Mechanical Strength and Microstructure of Green Ceramic Bodies.* Ceramica Acta. (6) 5-19, 1996.

feed and the pressing pressure. The maximum granule size must be limited to obtain a high mechanical strength in the tile. A higher pressing pressure may produce a higher strength when the pressing pressure does not exceed that causing laminations. Extremely large granules in the spray-dried powder tend to be irregular in shape and of variable density. The granulated feed should be screened to control the maximum granule size at about 500 μm. Also the size distribution of the larger 50% of granules should be controlled. The solution to maintaining a high compact density and strength without laminations is to maintain a well-controlled granulated feed. Surface cracks and edge chips also reduce pressed strength.

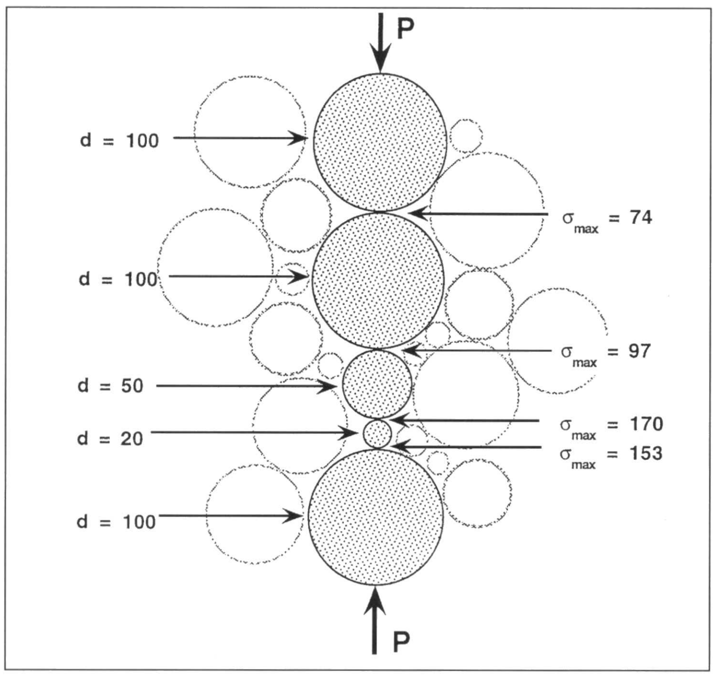

Figure 14. *Calculated contact stresses between elastic granules of different size (d in microns).*

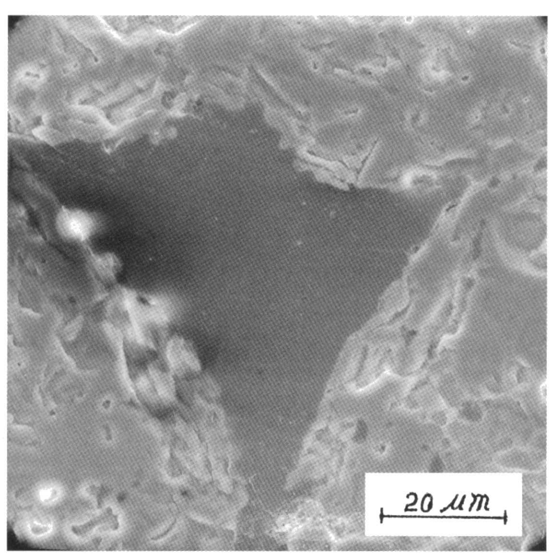

Figure 15. *Scanning electron micrograph showing a triangular defect (intersticie) between large granules after densification on firing.*

A region in the tile of lower pressed density will probably be of lower strength, and exhibit higher shrinkage on firing producing shape distortion. Density and strength uniformity depend on uniform filling, packing, deformation, and joining of granules. A non-uniform die-wall friction can produce density differences in the tile. The greater friction near corners may retard filling, reduce the pressing force, and produce a slightly higher porosity in a corner region. The pressure across the pressing surface will be lower near the center than near the edge. Dies must be well polished to minimize friction.

DIRECTIONS FOR PROCESS IMPROVEMENTS

Ceramic tile manufacturing has improved significantly during the recent decade and the understanding and control of process steps has greatly improved. Directions for improvements appear in four areas:

1. Process control improvements. Dimensional control, which becomes more demanding for larger tile, requires improved uniformity of the green bulk density. Control to $< \pm 0.02$ g/cm^3 is now a requirement for vitrified tile [2]. Both granule flow for die filling and granule deformation for compaction are sensitive to moisture content. Continuous control of the moisture content of the granule feed within very narrow limits is extremely important.

2. Improvement of granulate prepared using the dry preparation process. The dry preparation process enables lower running and production costs. However, the quality of the granulate feed and fired tile microstructure is reported to be sufficient only for downscale products[2]. Improved control of raw materials used and engineering improvements in processing machinery for granulation may enable preparation of improved pressing feed material.

3. Improve characteristics of spray-dried granules. The dependence of the characteristics of the granulate feed on slurry and spray-drying parameters is now much better understood in scientific terms. This knowledge should soon be translated into engineering practice.

4. Additives to improve process performance and/or reduce process sensitivity. Additives in an amount of 0.1 - 5% are used routinely for improving the pressing behavior and green strength of non-clay ceramics. A minor amount of an inorganic additive such talc that has very smooth surfaces and is hydrophobic may possibly improve both granule flowability and granule compaction. Talc in a colloidal form with plate-like particle shape would be most effective. The relatively high cost of an organic additive and the very short time for its burnout on firing the tile greatly limits the choice and amount of an organic processing additive for improving pressing and/or compact strength. Vinyl binders with a -C-C-C- backbone and cellulose binders are slow to burnout. A binder such as polyethylene oxide that burns out rapidly in air via. an "unzipping" of the C-O-C- linkage may be a potential binder candidate. Organic lubricants such as a stearate when added in an amount < 0.1% are effective in the pressing of alumina ceramics. These may be added into the slurry or tumbled with the granules to form a thin coating on the granule. Without a binder, a lubricant may reduce the green strength of a pressed clay ceramic.

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[2] NEGRE, F., AND SANCHEZ, E. *Advances in Spray-Dried Powder Processing for Tile Manufacture.* pp. 169-181 in Science of Whitewares. Edited by V. Henkes, G Onoda, and W. Carty. The American Ceramic Society, Westerville, OH USA. 1996.

To conclude, product competition provides a driving force for continued improvements in the tile quality. Final tile quality depends importantly on the processing before firing. Needed process improvements can benefit from a more scientific understanding of process steps and from improved process controls. Continued improvements in tile manufacturing appear probable.