# MEASUREMENT OF PARTICLE STRENGTH IN ALUMINA POWDERS USING ULTRASOUND

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**Abstract**

The strength of alumina is consistently rated as one of the most important product quality parameters. Particle strength is difficult to quantify and not well understood, which leads to inadequate control of fines generation, a costly problem for industry. CSIRO is developing a method and apparatus for the measurement of particle strength in powder beds using ultrasonic velocity measurements. It is considered to have several advantages over the current method for measuring alumina strength which is a modification of the Forsythe-Hertwig attrition index (AI) method, and has the potential to be reproducible, sensitive across the range of particle strengths encountered in alumina samples, quick to perform and independent of particle size.

Tests using the ultrasonic technique have been conducted on a range of calcined and uncalcined alumina samples to demonstrate empirically the relationship between ultrasonic measurements and particle strength. Good correlations between ultrasonic measurements and strength as measured by the breakage rate method were obtained with correlation coefficients in the range 0.8-0.9. To determine if there is any chance of measuring likely product strength before calcination, a correlation was made between the calcined breakage rate and uncalcined measurements of velocity and density, reasonable correlation was obtained with correlation coefficients in the range 0.75-0.8, these correlations were heavily dependent on the bulk density measurement. The size independence and non-destructive nature of this method of particle strength measurement were also established. The results of these tests demonstrate that the strength measurement apparatus and method is sufficiently developed for a large-scale assessment on alumina samples. Modification and automation of some aspects of the ultrasonic measurement procedure are expected to provide improvements in repeatability.

## 1. Introduction

In the Alumina industry vast quantities of alumina powder are produced by crystals grown in the Bayer process and calcined to meet size and purity standards. Strength of these powders is consistently rated as one of the most important product quality parameters as low strength can lead to excess fines being generated through attrition wear from handling. The inability to control fines generation in alumina refineries leads to a range of costly problems for producers including: excess fines leading to less product within specification, increased capital costs for fines handling equipment, increased loads on classification, increased recycling of material, reduced supersaturation and residence times in the precipitation stage, significant health hazards to personnel, and overall operational stability problems.

The paper "Alumina Fines' Journey from Cradle to Grave" (Chandrashekar 2005), describes alumina "dustiness" as possibly the highest priority concern for aluminium smelter operation. In the smelters, excess alumina fines generation produces increased particulate and fluoride emissions in the pot room. The paper suggests that size measurement at the "gate" of the smelter is an insufficient parameter as half the fines measured at the pot are generated within the smelter itself, and further emphasises the need for improvements in the understanding and measurement of alumina strength. To quote from the paper: "Of particular importance is the need to shift our focus to alumina strength rather than size. There is little point in the refinery minimising the –45 µm content of the alumina shipped to the smelter, only for the smelter to attrite the alumina during internal transfers. The challenge for researchers is to improve our understanding of strength and to develop a test which can be used for routine analyses."

The current method for measuring calcined and uncalcined alumina strength is a modification of the Forsythe-Hertwig attrition index (AI) method which is described in the following section. CSIRO has developed an alternative method and apparatus for the measurement of particle strength in powder beds.

This technique involves measuring the mean strength of the individual particles or pellets ultrasonically by measuring the sound speed. The ultrasonic technique is being developed because it has the potential to fulfil the criteria for a 'successful measurement technique for adaptation to an on-line system' as defined in an earlier AMIRA project (Illievski 2000). These criteria include:

- A reproducible and unique measure of 'toughness" i.e. materials with obvious differences in toughness characteristics should have different values
- Sufficient sensitivity to differentiate between a range of laboratory and refinery calcined and uncalcined alumina samples
- The technique has some physical meaning, which preferably would provide information on fines generation and breakage mechanism
- The technique is simple and quick to perform.

The ultrasonic technique also has the potential advantages of being size-independent and non-destructive.

## 2. Methods of Strength Measurement

The current method used for measuring alumina and uncalcined alumina strength is a modification of the Forsythe-Hertwig AI method (Benrose 1987). In this technique the –45 micron mass fraction is measured before and after treatment in a fluidised bed unit. The limitations of this method are high sensitivity of the measured AI to the operating parameters of the fluidised bed unit, and size dependence of the AI. Significantly different products can produce the same or similar measured AI values and in some cases a product with less desirable breakage and attrition properties can even have a lower AI. In addition to these drawbacks, the current method is impossible to adapt for on-line measurement should that be desired.

In contrast to the Forsythe-Hertwig AI method, the ultrasonic technique is directly related to a basic physical property.

Sound is the propagation of a physical disturbance, such as the compression of a medium. In bulk solids the sound speed depends on two factors, the density and elastic modulus of the material (Digby 1981). The elastic modulus is related to the force required to microscopically deform the solid, which has an intuitive link "strength" or "resilience" of the material. In the case of composite materials, such as pellets, this modulus can be shown to have a direct theoretical connection to the strength of the bonds holding the composite particle together (Garcia 1999). An attrition process knocks off bits of a particle and so should also be related to how well a particle such as alumina powder, which is formed principally by an agglomeration process, is stuck together. The work described in this paper has been directed at demonstrating this relationship empirically in alumina powder.

Sound speed measurements have been made in sintered powders to measure the degree of neck formation between powders and hence the strength of the sinter (Garcia 1999). However, to the authors' knowledge, sound speed measurements of unbonded particles with the intention of determining particle strength have not been performed previously. Measurement of an ensemble of particles presents problems of interpretation that are discussed in the next section, but the resultant sound speed or Young's modulus measurement can be compared to the more traditional measurement of alumina strength, i.e. attrition tests. For alumina the density of the particles is approximately uniform so either the modulus or the velocity can be used interchangeably.

## 3. Sound Transmission Through Granular Solids

Sound transmission through granular solids has been studied in a number of applications. For the case of low frequency sound transmission there is a well-developed body of literature to describe sound velocity, for example on seismic waves passing through sand and gravel (Digby 1981), sonar from sea beds (Buckingham 1997) and sound transmission in sinters (Garcia 1999). Sound attenuation, being much more difficult to describe, has received less attention (Jia 1999).

The ultrasonic method developed by CSIRO requires attenuation to be small enough to allow for sound speed measurement; measurement frequencies are selected based on grain size to minimize scattering. This discussion is focused on using velocity measurement to determine particle strength.

Much of the physics of sound transmission in granular solids can be understood relatively simply in terms of contact between spheres. In a loose aggregate there is essentially no transmission of sound waves through the particles as there is no resistance to small displacements of the particles. Pressure is required to make solid contacts between particles and allow for sound transmission. Figure 1 shows two elastic spheres under a force that results in a contact area of radius *a*. The contact radius *a*, is caused by the force, and when averaged over many spheres of uniform radius *R* in a powder, increases with pressure *P*.

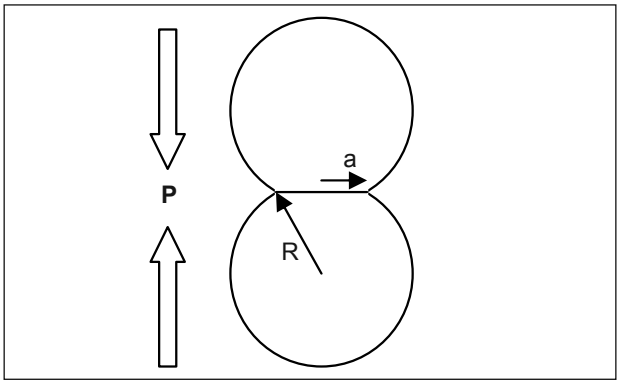


Figure 1. Two spheres of radius *R* in contact under pressure *P* have a contact area of radius *a*.

A comprehensive examination (Digby 1981) of the theory with spheres in a fixed random packing gives the longitudinal sound velocity *V* as

$$V = \frac{0.39}{\sqrt{\rho_b}}\left( (KE)^{1/3}\left(\frac{1}{1-\nu_b}\right)^{1/3}\left(\frac{1}{1-\alpha}\right)^{1/6}(P)^{1/6}\right) \qquad [1]$$

where $\rho_b$ is the density of the particles, K is the average number of contacts for each sphere (which for a random packing is 8.84), $\alpha$ is the voidage fraction and $\nu_b$ is the Poisson's Ratio of the bulk material. The main assumptions made in these calculations are that the wavelength of the sound is much larger than the particle radius, that the radius is constant for all the particles, and that pressure is insufficient to significantly deform the particles.

Equation 1 provides a list of the factors needed to be taken into account in a measurement of the Young's Modulus E or sound speed of the particles in a powder. K represents the configuration of the particles, which is difficult to measure. However, standardisation of sample preparation does produce relatively consistent levels of compaction for each of the materials being tested. The bulk density of the particle bed (to obtain voidage fraction) and the pressure need to be measured when the velocity measurement is made. The specific gravity and Poisson's Ratio of the individual particles are assumed to be known and to be fixed properties of the material. It can be seen from Equation 1 that there is no direct dependence on particle size for sound velocity.

The theory is essentially the same for the shear wave velocity except the Young's Modulus E is replaced by the Shear Modulus G in Equation 1. The shear modulus, as the name suggests, relates to the response perpendicular to the applied pressure in Figure 1. It is possible that shear response may be more closely related than longitudinal velocity to dusting as most impacts are glancing.

## 4. Experimental Measurements

The apparatus for the sound velocity measurement is shown in Figure 2. The cylindrical sample holder contains the receive transducer in its base. During measurement the sample holder is fixed in position on the apparatus. The powder to be measured is placed in the sample holder and a fixed pressure is applied to the sample by lowering the ram. Between the top of the sample and the ram is the transmit transducer which is pressed onto the sample when the ram is lowered. A sound pulse is then transmitted through the sample from top to bottom and the time of flight is measured. Figure 3 shows schematics of transmit and receive transducer pairs used for longitudinal and shear measurements.

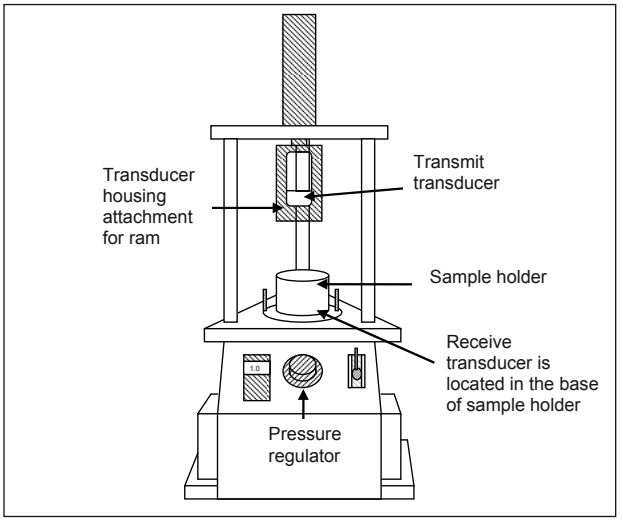


Figure 2. Schematic of measurement apparatus. Note: longitudinal transducer pair is shown in this figure. Schematic of shear transducer pair for use in the same apparatus (with transducer housing attachment for ram removed) is shown in Figure 3B.

The internal diameter of the sample holder is 60mm. The external diameter of the top transducer piece (for both longitudinal and shear measurements) is 56mm to avoid powder becoming stuck between the top transducer piece and the wall of the sample holder. If the powder becomes stuck in this way, then the full pressure will not be applied to the powder being measured.

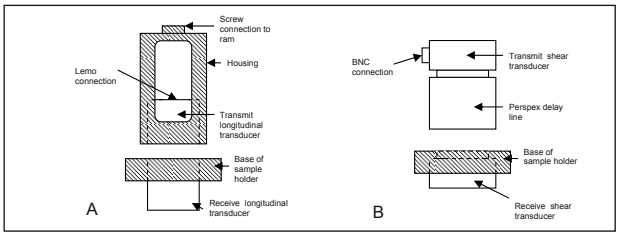


**Figure 3. Schematic of transmit and receive transducer pairs for (A) longitudinal measurement, and (B) shear measurement.**

### 4.1 Equipment

Longitudinal measurements were made at 0.5 MHz using Krautkramer non-destructive testing transducers and an Accutron 1010PR pulser / receiver. For the longitudinal configuration of the apparatus (Figure 3a), the top transducer was attached to the ram which was lowered into the sample holder to apply pressure to the sample being measured. The transducer width for the longitudinal transducers was 40mm, which was equivalent to an application of sound to 0.44 of the surface area of the sample.

Shear measurements were made at 100kHz using Panametrics V1548 transducers. For shear configuration of the apparatus (Figure 3b) the top transducer was attached to a delay line which was placed on top of the sample inside the holder and pressure applied to the transducer top piece using the ram. The top transducer had a Perspex delay line of 28mm thickness glued to the transducer front face. The signal was generated using an Agilent Signal generator and an ENI RF Power Amplifier. The transducer width for the shear transducers was 45mm, which was equivalent to an application of sound to 0.56 of the surface area of the sample.

The received ultrasonic signal was digitised and averaged on a Tektronix TDS1002 oscilloscope.

### 4.2 Measurement Procedure

The measurement procedure was standardised for longitudinal and shear measurements on a range of materials to enable comparison of results. The measurement procedure is as follows:

- With the sample holder in position, the sample powder is sifted, weighed and poured into the sample holder. The sample is then levelled by shaking the sample holder horizontally. Once the sample is levelled the transmit transducer is rested on the powder and settled in a horizontal position
- A pressure of 2 bars is applied to the pneumatic ram for a standard velocity measurement. The actual pressure applied to the sample is 1 bar as the internal area of the sample holder is twice the ram area
- To induce closer packing in the sample, the side of the sample holder is tapped using a hammer
- Measurements of sound transmission are made as the sample settles. A signal of at least 800mV Pk-Pk was the minimum amplitude of received signal considered acceptable for any measurement
- Time delay is measured using the oscilloscope as the first negative baseline crossing of the signal. The shortest time delay measured during tapping was accepted as the time delay measurement for that sample. The signal used for the measurement was an average of 16 pulses. Displacement of the top transducer (which equates to the thickness of the powder) was measured at the shortest time delay.

The total time to complete the procedure for one sample is approximately 10 minutes. Results from this procedure are used to determine the bulk density and speed of sound. These are put into Equation 1 and used to calculate the Young's modulus or Shear modulus or equivalently the longitudinal or shear velocity of the powder particles.

### 4.3 Size Independence of Procedure

Experiments were conducted to test the size independence of the measurement procedure by measuring sound velocity in different grades of glass ballotini. These grades have narrow particle size distributions. The ballotini grades were chosen to cover the size range present in alumina powders. The shape of the ballotini is spherical so the packing configuration for this material is simple. The ballotini can be used to test the size independence of the strength measurement procedure under the assumption that the strength is uniform across ballotini grades.

Three measurements of longitudinal ultrasonic velocity were performed for each of the ballotini grades on 25 gm samples at 0.5 MHz; the results are shown in Table 1. The measured velocities can be seen to be essentially the same for all the ballotini grades independent of the particle size. The standard deviation is higher than the estimated measurement error of approximately 3 m/s. This is due to difficulties in presenting the sample in the same state for each measurement.

**Table 1: Velocity measurement in Glass Ballotini at 0.5 MHz**

| Grades of Glass Ballotini | D50* | Mean Velocity (m/s) | Standard Deviation (m/s) |
|---|---|---|---|
| **AH** | 70 | 700 | 10 |
| **AE** | 115 | 702 | 29 |
| **AC** | 191 | 687 | 21 |

* D50 is the median particle size

## 5. Measurement of Alumina Powders

### 5.1 Demonstration of the Non-destructive Nature of the Procedure

Subsamples of alumina powder were taken before and after a test run of the measurement procedure; these samples were sent for size distribution analysis by optical diffraction. For the purposes of this demonstration, the material was vibrated mechanically during the test run well in excess of the level required for alumina measurement. Results of the size distribution analysis are shown in Figures 4a, 4b and Table 2. It can be seen that there is essentially no difference between the size distributions regardless of whether the subsamples were collected before or after the test, showing that no damage is caused to alumina by the measurement procedure.

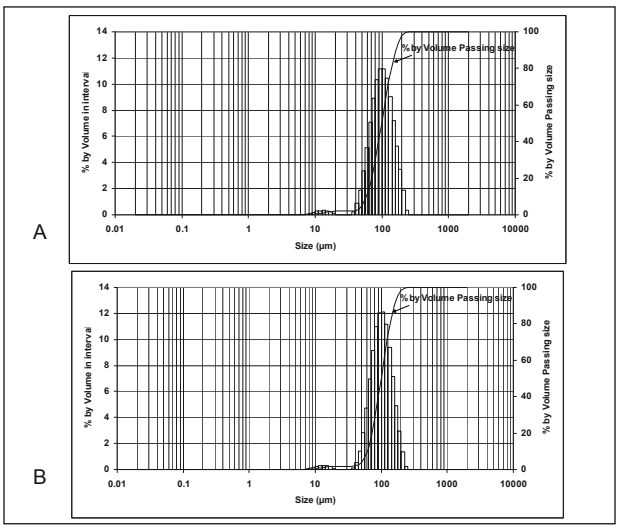


Figure 4: Size distribution of alumina (a) before measurement procedure and (b) after measurement procedure. *Note: Size cut-point results from the graphs are summarised in Table 2.*

Table 2. Size measurement of Alumina before and after agitation

| Size Cut-point | Particle Size (μm) | |
|---|---|---|
| | Before Measurement (A) | After Measurement (B) |
| D(0.1)* | 58.679 | 61.317 |
| D(0.5) median particle size | 99.196 | 99.577 |
| P80 (80% by volume passing size) | 138.29 | 135.41 |
| D(0.9) | 162.09 | 157.2 |

* Note: D(0.1) refers to the point on the size distribution "% by Volume in interval" where 10% of solids are below that particle size and 90% are above it.

### 5.2 Comparison of Ultrasonic and Breakage Rate Measurements

A set of uncalcined and calcined alumina samples were used to test empirically the relationship between ultrasonic measurements and strength as measured by breakage rate. The calcined alumina samples were produced by calcining (heating) a portion of each of the uncalcined samples. This process drives off water from the particles and leads to a porous, more friable powder. Breakage rate data for these alumina samples was taken from Illievski (2000).

Sound velocity measurements were made in the alumina samples using the method described above. Measurements were conducted using both shear and longitudinal ultrasound to determine whether shear velocity is more closely related to the results of attrition testing. It was found that the measured sound velocity could vary quite widely during consolidation of the powder therefore the bulk density of the powder was included in the analysis of the results.

Breakage rate measurements are performed by measuring the particle size distribution of an alumina, placing it in a fluidised bed for a set period and re-measuring the size distribution. This procedure is repeated a number of times. An index, *k1*, is developed from these measurements to indicate the rate of change of the size distribution. The breakage rate measurements provide a rough guide of the relative strength of the alumina samples which can be compared to ultrasonic measurements. A higher breakage rate implies a weaker powder.

Figure 5a shows the results of a linear two-term correlation between longitudinal ultrasonic velocity and bulk density measurements with the breakage rate parameter *k1* for the uncalcined and calcined samples. Figure 5b shows the procedure repeated with shear acoustic measurements. The correlation coefficients and terms used in the correlations are given in Tables 3 and 4. The results of two correlations are shown, one with the bulk density term and one without.

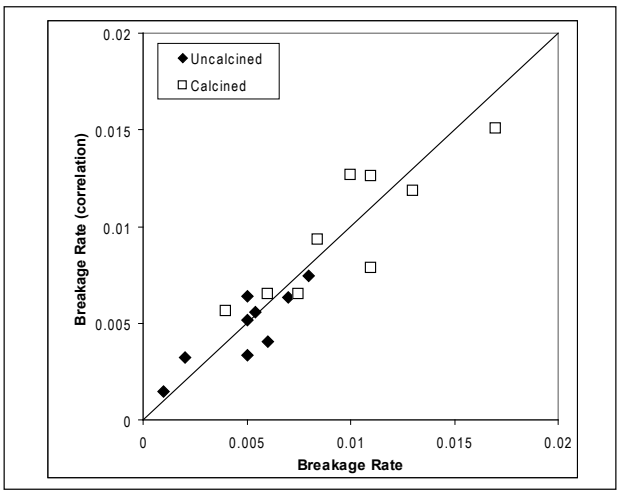


Figure 5a. Comparison of predicted to actual breakage rate for calcined and uncalcined alumina using **longitudinal** ultrasonic measurements.

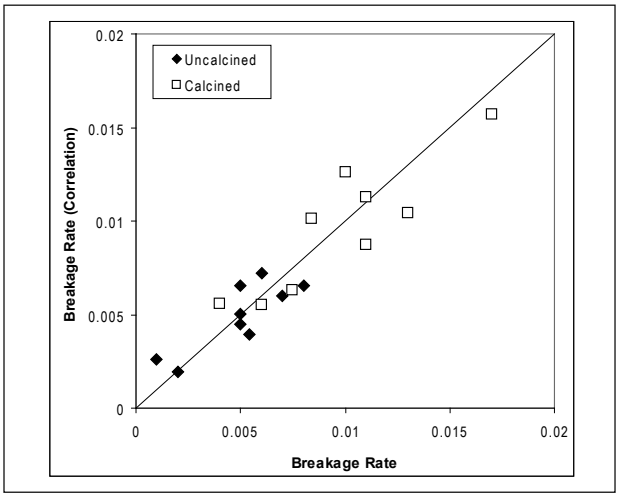


Figure 5b. Comparison of predicted to actual breakage rate for calcined and uncalcined alumina using **shear** ultrasonic measurements.

Table 3. Correlation values for strength inferred from longitudinal ultrasonic measurements to breakage rate

| Correlations for Longitudinal US | Uncalcined correlations | | Calcined Correlations | |
|---|---|---|---|---|
| | Bulk Density Term | No Bulk Density Term | Bulk Density Term | No Bulk Density Term |
| Correlation Coeff. | 0.85 | 0.644 | 0.88 | 0.88 |
| Velocity term | $-2.6\ 10^{-5}$ | $-2.5\ 10^{-5}$ | $-13.7\ 10^{-5}$ | $-11.7\ 10^{-5}$ |
| Bulk density term | -0.024 | | 0.0048 | |

Table 4. Correlation values for strength inferred from shear ultrasonic measurements to breakage rate

| Correlations for Shear US | Uncalcined correlations | | Calcined Correlations | |
|---|---|---|---|---|
| | Bulk Density Term | No Bulk Density Term | Bulk Density Term | No Bulk Density Term |
| Correlation Coeff. | 0.83 | 0.713 | 0.89 | 0.88 |
| Velocity term | $-5.5\ 10^{-5}$ | $-4.32\ 10^{-5}$ | $-6.9\ 10^{-5}$ | $-7.2\ 10^{-5}$ |
| Bulk density term | -0.01 | | 0.00105 | |

Reasonable correlations exist for both shear and longitudinal ultrasound to breakage rate for both calcined and uncalcined alumina, as shown in Figures 5a and 5b. For calcined alumina the breakage rate correlations have limited dependence on the bulk density measurement. In uncalcined alumina some dependence is present though still of much less importance than the sound velocity measurement. In general, the lower the breakage rate (i.e. the stronger the material) the faster the ultrasonic velocity; this leads to negative velocity terms in the correlation equations. There was no significant difference between the shear and longitudinal measurements provided the bulk density term was included. These results provide evidence that a measure of particle strength can be related to the velocity of sound in alumina.

To determine if there is any chance of measuring likely product strength before calcination, a correlation was made between the calcined breakage rate and uncalcined measurements of velocity and density. This correlation for longitudinal and shear measurements respectively is shown in Figures 6a and 6b. The values for the correlations and the terms used are given in Table 5.

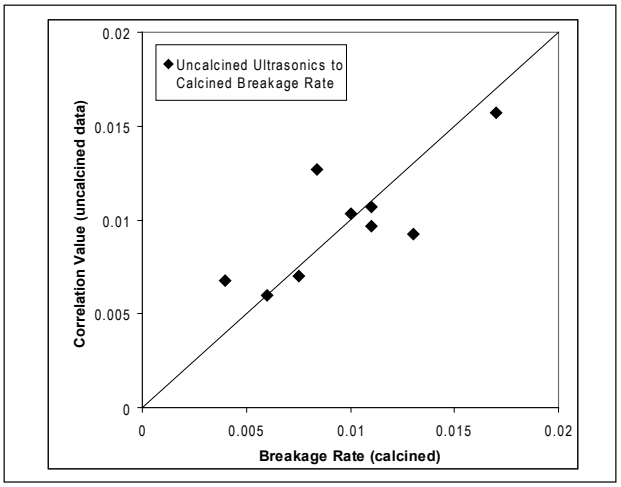


Figure 6a. Uncalcined **longitudinal** velocity and bulk density measurements correlated to calcined breakage rate.

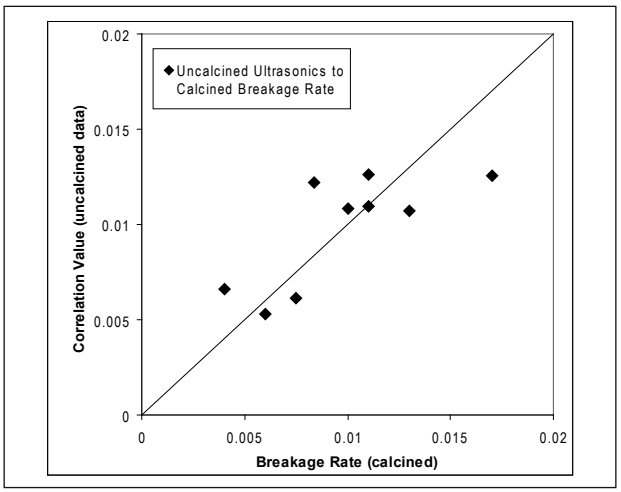


Figure 6b. Uncalcined **shear** velocity and bulk density measurements correlated to calcined breakage rate.

**Table 5. Correlations values for strength inferred from uncalcined ultrasonic measurements to calcined breakage rate.** ***Note: The last column is the correlation between the uncalcined and calcined breakage rate measurements.***

| Correlation | Longitudinal Correlations | | Shear Correlations | | Bulk Density Correl'n | Breakage Rate |
|---|---|---|---|---|---|---|
| | Bulk Density Term | No Bulk Density Term | Bulk Density Term | No Bulk Density Term | | |
| Correlation Coeff. | 0.80 | 0.044 | 0.754 | 0.336 | 0.646 | 0.43 |
| Velocity term | $1.44\ 10^{-6}$ | $2.94\ 10^{-6}$ | $-5.5\ 10^{-5}$ | $-3.57\ 10^{-5}$ | | |
| Bulk density term | -0.048 | | -0.0285 | | -0.048 | |

## 6. Discussion

Good correlations exist for both longitudinal and shear ultrasound to the breakage rate of both calcined and uncalcined alumina as shown in Figures 5a and 5b. These correlations show little or no dependence on bulk density measurement, especially in the case of the calcined alumina measurements. The general trend between breakage rate and ultrasonic velocity is that lower breakage rates (i.e. stronger materials) are associated with faster ultrasonic velocity. This leads to negative velocity terms in the correlations shown in Tables 3 and 4. The bulk density term has an opposite sign for the uncalcined and calcined correlations shown in Tables 3 and 4. At this stage shear and longitudinal techniques will continue to be evaluated in parallel as it is unclear which will ultimately prove the most useful. For prediction of calcined alumina strength from uncalcined alumina measurements the bulk density is far more important, with correlations based only on ultrasonic velocity having poor correlation coefficient as shown in Table 5, whilst that based on bulk density alone demonstrates a significant level of correlation.

These tests demonstrate that our strength measurement apparatus and method is sufficiently developed to embark on a large-scale assessment of alumina samples. To provide a comprehensive evaluation of the measurement technique it is proposed that samples be collected from a number of points along the alumina production process as material progresses from uncalcined alumina production stage right through to entry in a cell at the smelter. Lab analysis of these samples off-line would involve size distribution by optical diffraction and ultrasonic measurements using the strength measurement apparatus. This allows for comparison of the ultrasonic measurements against a relevant industry performance measure, viz., the amount of fine material produced by handling.

The measurement technique can be further improved, especially by increasing the reproducibility of measurements. To achieve this the apparatus is being modified to have the agitation and ultrasound pulse/receive/measurement electronics automated. The first priority is the sample agitation as most of the measurement variation observed appears to be due to sample presentation and compaction conditions.

## 7. Conclusions

CSIRO has developed a new technique for the determination of particle strength in powder beds from measurement of ultrasonic velocity. The method is considered to have several potential advantages over the Forsythe-Hertwig AI method in that it is independent of particle size and is directly related to a physical property.

The strength measurement apparatus has been tested on a range of calcined and uncalcined alumina samples and the relationship between ultrasonic measurements and strength was demonstrated empirically:

- Correlations of both shear and longitudinal ultrasonic velocity with breakage rate provide correlation coefficients in the range 0.8-0.9. This is a good result given the limits of the breakage rate measurement in assessing strength
- Predictions of calcined alumina behaviour from uncalcined alumina properties depend more on bulk density measurements than the velocity measurements and provide reasonable correlations with correlation coefficients in the range 0.75-0.8.

The strength measurement apparatus and method is sufficiently developed for a test of its applicability through a large-scale assessment of alumina samples. Significant improvements in repeatability are expected when aspects of the measurement procedure are improved and automated.

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