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# Non-destructive characterization of zirconia-toughened alumina ceramics using ultrasonics

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Ultrasonics has been used for the characterization of sintered zirconia-toughened alumina (ZTA) ceramics. The variation of ultrasonic velocity with volume fraction of zirconia in an alumina matrix was studied. Variation of ultrasonic velocity with volume fraction of $ZrO_2$ content was analysed in terms of a second-degree polynomial in the volume fraction of $ZrO_2$. The elastic moduli and Poisson's ratio evaluated from the longitudinal and shear velocities have also been described in terms of a similar equation. The observed variation in velocity and moduli with porosity has been compared with the predicted values based on elasticity theory.

## 1. Introduction

In the last few decades, considerable interest has been developed in the non-destructive ultrasonic characterization of ceramics, electrical porcelain and other porous materials, for their physical and elastic properties [1–5]. Clay based ceramics have also been investigated to find the effect of compaction parameters on the sintered properties [6] and establish the correlation between the physical properties and pore volume fraction [7]. In recent years studies have been carried out on ultrasonic characterization of multi-phase composites such as graphite/epoxy composites [8–12], aluminium/aramid-epoxy composite [13], multidirectional composite laminate [14], etc. Lu *et al.* [15] evaluated silicon carbide-reinforced aluminium metal matrix composites and tried to establish the effect of porosity on the ultimate physical and elastic properties, while Jagnoux *et al.* [16] studied the interface in single SiC fibre-reinforced aluminium composites using a similar technique. However, it seems that ultrasonic characterization of particulate-toughened ceramics, to the best our authors knowledge, has not received much attention. An attempt has therefore been made in this work to characterize zirconia-toughened alumina, to evaluate the elastic constants, and find the correlation between physical properties and volume fraction of zirconia content.

## 2. Experimental procedure

### 2.1. Specimens

Powders of alumina (XA-16, USA) and varying amounts of zirconia, partially stabilized with 3 mol % $Y_2O_3$ (3Y-TZP, Tayosoda Corporation, Japan) were milled in a planetary ball mill in isopropanol medium to prepare the basic composite powders. The particle size and the chemical analysis of the powders are given in Table I. The mixture was subsequently dried and calcined at 700 °C. The calcined product was reground in isopropanol and polyvinyl alcohol was added during grinding. These powders were uniaxially pressed into plates of $25 \times 25 \times 5\ mm^3$ dimensions at 120 MPa. The green compacts were sintered at 1550 °C for 2 h in air. The sintered samples were subsequently surface ground and polished with a lap-master to maintain the surface parallelity of the faces within 1 μm.

The bulk density, ρ, of the sintered body of various ZTA compositions was measured by the water displacement method while their theoretical density, $\rho_0$, was calculated from the available atomic weights and unit cell dimensions. The total porosity was then calculated from the relation

$$P = 1 - \rho/\rho_0 \quad (1)$$

where $P$ is the pore volume fraction.

### 2.2. Ultrasonic velocity measurement

The ultrasonic velocity (transverse and longitudinal) was measured using the ultrasonic Tester USIP 12 (Kraut Kramer) interfaced with a 100 MHz oscilloscope (Philips model 3350). The centre frequency of the transducer (10 mm diameter) was 15 and 5 MHz for longitudinal and transverse waves, respectively. The pulse–echo method was used for calculating the velocity. The transient time could be measured within an accuracy of ± 5 ns.

## 3. Results

### 3.1. Ultrasonic velocity

Table II gives the ultrasonic velocity of samples of different $ZrO_2$ content together with the estimated

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TABLE I Chemical analysis of alumina and 3Y-PSZ powders

| Constituents (wt %) | Alumina (grade XA-16 supplied by Alcoa) | 3Y-PSZ (supplied by Tayosoda) |
|---|---|---|
| $Y_2O_3$ | – | 5.07 |
| $Al_2O_3$ | Balance | < 0.005 |
| $ZrO_2$ | Trace | Balance |
| $SiO_2$ | 0.02 | < 0.002 |
| $Fe_2O_3$ | 0.006 | < 0.002 |
| $Na_2O$ | 0.07 | 0.015 |
| Loss on ignition | – | 0.71 |

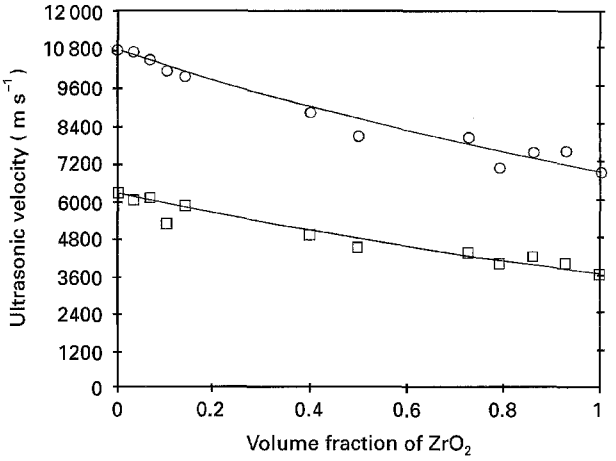


*Figure 1* Variation of (○) longitudinal and (□) transverse velocity with volume fraction of $ZrO_2$ in sintered ZTA ceramics. (——) Fitted Equations 10 and 11.

porosity values. It may be seen from the table that the porosity of the sample is less than 8%, though they differ from sample to sample.

In a recent review of the data available on porous polycrystalline materials, Roth *et al.* [17] showed that the ultrasonic velocity, $V$, variation with porosity, $P$, is given by the relation

$$V = V_0 (1 - P) \tag{2}$$

where $V_0$ is the velocity for zero porosity. Thus, for comparison of ultrasonic velocity variation with volume fraction of $ZrO_2$ zero porosity values are obtained from the above relation using the measured velocity and porosity data. Those values are also given in Table II. Fig. 1 presents the plot of longitudinal, $V_l$, and transverse, $V_s$, ultrasonic velocity against volume fraction of zirconia.

The measured values were fitted to an equation of the form

$$V_l = V_{l0} (1 - a\phi + b\phi^2) \tag{3}$$

$$V_s = V_{s0} (1 - a_1\phi + b_1\phi^2) \tag{4}$$

where $V_{l0}$ and $V_{s0}$ are the ultrasonic velocities in pure alumina, $a$, $b$, $a_1$, $b_1$ are constants and $\phi$ is the volume fraction of zirconia.

The above relations were derived from Hashin's relation from elastic moduli of a two-phase material [18]. For a two-phase material, Hashin gave a good approximation of the shear modulus

$$G^*/G_m = 1 + \frac{A\ (D-1)\phi}{1 + C\ [D - (D-1)\phi]} \tag{5}$$

where $G^*$ is the shear modulus of the two-phase system, $G_m$ is the shear modulus of the matrix, $A$, $C$, $D$ are constant for a particular combination of the matrix and inclusion, $A$ and $C$ are functions of Poisson's ratio of the matrix, and $D$ is the ratio of shear modulus of inclusion and the matrix.

Equation 5 can be further simplified to the form

$$G^*/G_m = \frac{1 - C_1\phi}{1 - B_1\phi} \tag{6}$$

where $C_1$ and $B_1$ are again functions of the constants $A$, $C$ and $D$, because

$$V_s^2 = G^*/\rho^* = \frac{G^*}{\rho_m\ (1 - \phi) + \rho_1\phi} \tag{7}$$

where $\rho_1$ and $\rho_m$ are the densities of the inclusion and the matrix, respectively.

TABLE II Ultrasonic velocities of ZTA ceramics

| $ZrO_2$ (vol %) | Density (g cm$^{-3}$) | Porosity (%) | Measured long. vel. (m s$^{-1}$) | Measured trans. vel (m s$^{-1}$) | Longitudinal vel. for zero porosity (m s$^{-1}$) | Transverse vel. for zero porosity (m s$^{-1}$) |
|---|---|---|---|---|---|---|
| 0 | 3.95 | 1.00 | 10724.1 | 6242.4 | 10832.4 | 6305.5 |
| 3.3 | 4.04 | 1.20 | 10609.9 | 6000.0 | 10740.9 | 6074.1 |
| 6.8 | 4.13 | 1.20 | 10374.7 | 6052.6 | 10500.7 | 6126.1 |
| 10.4 | 4.22 | 0.90 | 10053.1 | 5241.4 | 10148.5 | 5291.1 |
| 14.1 | 4.28 | 1.40 | 9821.1 | 5789.5 | 9958.5 | 5870.5 |
| 39.6 | 4.68 | 7.20 | 8158.4 | 4564.3 | 8791.4 | 4918.4 |
| 49.6 | 4.90 | 7.90 | 7403.6 | 4138.2 | 8038.7 | 4493.2 |
| 72.4 | 5.48 | 2.50 | 7638.3 | 4133.0 | 7834.2 | 4238.9 |
| 78.8 | 5.54 | 2.00 | 7017.2 | 3970.7 | 7160.5 | 4051.7 |
| 85.5 | 5.62 | 3.60 | 7219.0 | 4043.0 | 7488.6 | 4194.0 |
| 92.6 | 5.71 | 4.00 | 7238.0 | 3814.8 | 7539.6 | 3973.4 |
| 100 | 6.02 | 1.00 | 6815.5 | 3591.8 | 6883.7 | 3627.8 |

Combining Equations 6 and 7

$$V_s^2 = V_{s0}^2 \frac{(1 - C_1\phi)}{(1 - B_1\phi)(1 - f_1\phi)} \quad (8)$$

where $f_1$ is the density ratio between inclusion and matrix. Expanding the terms of Equation 7 binomially, and neglecting terms higher than second order in $\phi$, yields Equation 4. For longitudinal velocity, a similar relationship was also assumed.

Equations 3 and 4 were fitted to the data by regression analysis. The sum of squares, $Q$, was used as the measure of the goodness of the fit between the fitted equation and the data

$$Q = 1 - \sum_{i=1}^{n} (M_i - \hat{M}_i)^2 / \sum_{i=1}^{n} (M_i - \bar{M}_i)^2 \quad (9)$$

where $\hat{M}_i$ is the value calculated from the fitted equation for an appropriate $\phi$ value. $M_i$ and $\bar{M}_i$ are the measured values and mean value, respectively. For a good fit $Q > 0.95$; for $Q < 0.9$, the fit is poor. The equations that fit the data are:
longitudinal velocity

$$V_l(\mathrm{m\,s^{-1}}) = 10\,871.94\,(1 - 0.61\phi + 0.268\phi^2) \quad (10)$$

shear velocity

$$V_s(\mathrm{m\,s^{-1}}) = 6191.88\,(1 - 0.626\phi + 0.243\phi^2) \quad (11)$$

with $Q$ values of 0.98 and 0.95, respectively, for longitudinal and shear ultrasonic velocities, indicating a good agreement between the fitted equation and the data, the extrapolated values for ultrasonic longitudinal and shear velocities for pure alumina (i.e. $\phi = 0$) and pure zirconia (i.e. $\phi = 1$) work out to be 10871.9, 6191.9 and 7153.7, 3820.4 $\mathrm{m\,s^{-1}}$, respectively. These values compare favourably with the respective values of theoretically dense alumina for longitudinal and shear velocities of 10 845 and 6373 $\mathrm{m\,s^{-1}}$, and 9920 and 6350 $\mathrm{m\,s^{-1}}$, obtained by Spriggs [19] and Natarajan [20], respectively. In the absence of any reported values of theoretically dense zirconia ceramics with 3 mol% $Y_2O_3$, other extrapolated values obtained in this work for $\phi = 1$ could not be compared. However, Kandil *et al.* [22] reported the longitudinal and transverse ultrasonic velocities of $ZrO_2$ with 8 mol% $Y_2O_3$ as 7100 and 3060 $\mathrm{m\,s^{-1}}$, respectively. These values are in fair agreement with the present values of $ZrO_2$ with 3 mol% $Y_2O_3$.

Theoretically, the ultrasonic velocities of two-phase materials can be estimated from the relation

$$V_l = \left[\frac{K + 4/3G}{\rho}\right]^{1/2} \quad (12)$$

$$V_s = \left[\frac{G}{\rho}\right]^{1/2} \quad (13)$$

where $K$ and $G$ are the theoretically estimated bulk modulus and shear modulus of two-phase systems. The theoretical estimate of $K$ and $G$ can be obtained from the Hashin's elasticity theory which considers the change in strain energy in a loaded homogeneous body due to the presence of non-homogeneity. The theoretical values of the module were estimated using from the bounds of bulk modulus and the approximate expression for shear modulus. Equations 12 and 13 were then used for calculating theoretical velocity values. These estimated values, along with the experimental data, are presented in Fig. 2. The predicted values are in close agreement with the experimental data. In deriving these values, the bulk shear moduli of theoretically dense alumina were taken as 251.0 and 162.9 GPa, respectively. These values were estimated by Anderson *et al.* [21] from measured single-crystal elastic constants.

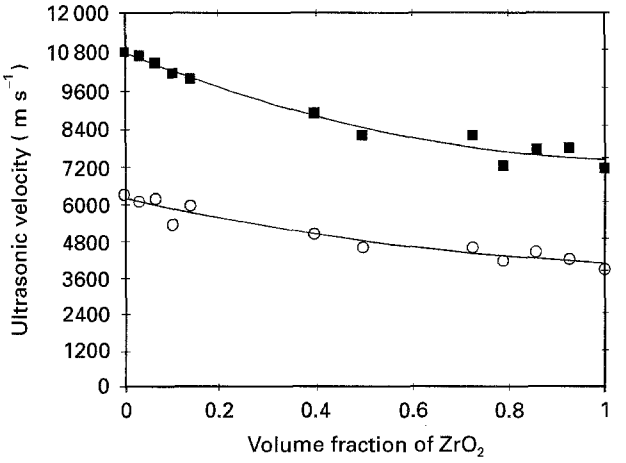


*Figure 2* Comparison of ultrasonic velocity variation in sintered ZTA ceramics with (——) the theoretically predicted values: (■) longitudinal, (○) transverse.

It may be noted here that the theory assumes a spherical shape of the inclusion. Thus the close agreement between theory and experiment indicates that zirconia grains are nearly spherial in shape. This is confirmed from the micrographs shown in Fig. 3 for volume fraction of zirconia samples.

## 3.2. Elastic moduli

The elastic moduli of the samples were calculated from the measured ultrasonic velocities using the equations:
Young's modulus

$$E = \frac{\rho\, V_s^2\, (3V_l^2 - 4V_s^2)}{(V_l^2 - V_s^2)} \quad (14)$$

shear modulus

$$G = \rho\, V_s^2 \quad (15)$$

Poisson's ratio

$$\nu = \frac{(V_l^2/2 - V_s^2)}{(V_l^2 - V_s^2)} \quad (16)$$

Fig. 4 presents the Young's and shear moduli plotted against the volume fraction of $ZrO_2$. Again, equations

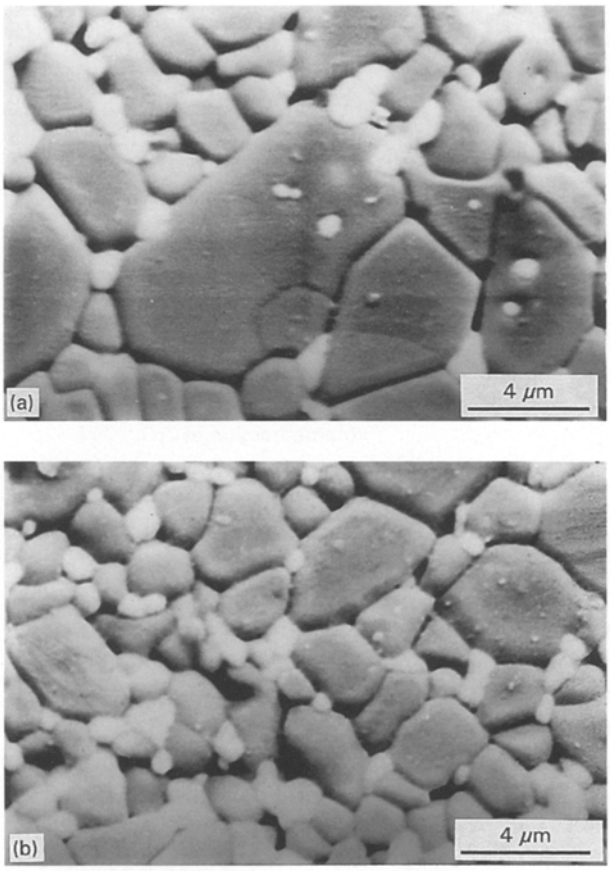


*Figure 3* Micrographs of ZTA ceramics (a) with 3.3% and (b) 10.4% $ZrO_2$ showing the nearly spherical shape of $ZrO_2$ grains.

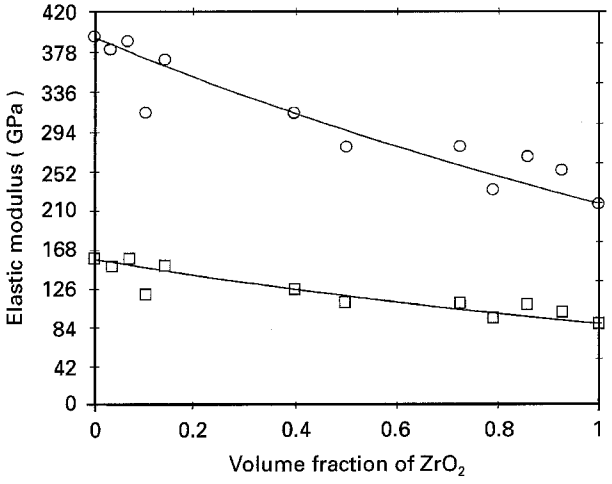


*Figure 4* Variation of elastic moduli of sintered ZTA ceramics with volume fraction of $ZrO_2$. (——) Fitted Equations 19 and 20, respectively: (○) Young's, (□) shear moduli.

of the form

$$E = E_m (1 - a\phi + b\phi^2) \quad (17)$$

and

$$G = G_m (1 - a_1\phi + b_1\phi^2) \quad (18)$$

were fitted to the experimental data by regression analysis. This yielded the relations

$$E\ (\text{GPa}) = 386.3\ (1 - 0.63\phi + 0.21\phi^2) \quad (19)$$

$$G\ (\text{GPa}) = 153.9\ (1 - 0.65\phi + 0.21\phi^2) \quad (20)$$

TABLE III Comparison of elastic moduli values reported in the literature

| Material | Young's modulus (GPa) | | Shear modulus (GPa) | |
|---|---|---|---|---|
| Alumina | 386.3 | Present work | 153.9 | Present work |
| | 402.0 | [23] | 160.6 | [21] |
| | 397.0 | [24] | 163.3 | [22] |
| Zirconia | 227.1 | Present work | 87.6 | Present work |
| | 209.0 | [25] | – | |
| | 207.0 | [26] | – | |

giving $Q$ values of 0.898 and 0.875, respectively. These equations are again plotted in Fig. 4 showing good agreement with the data.

The extrapolated value of Young's and shear moduli of pure alumina and zirconia are obtained as 386.3 and 153.9, and 227.14 and 87.6 GPa, respectively. These values, along with those reported in the literature, are compared in Table III.

Table III shows that the extrapolated values are in fairly good agreement with the values reported in the literature. The theoretical values of elastic moduli were also calculated using Hashin's theory. The calculated, as well as the measured, values are plotted in Fig. 5. The variation in moduli predicted by the theory agrees fairly well with the observed values. This again confirms the spherical nature of the $ZrO_2$ grains. A more sensitive test of the power of theory is a comparison between the bulk modulus and Poisson's ratio and values of these isotropic elastic constants calculated from the values of $E$ and $G$. This comparison is shown in Fig. 6. The closeness of these calculated values with the theoretical curve is remarkable considering the sensitivity of this calculation to the propagation of experimental errors. Particularly for Poisson's ratio, this sensitivity is magnified around low-volume fractions of zirconia due to the substraction of two almost equal terms.

## 4. Discussion

It has been observed that variation of ultrasonic velocity in zirconia-toughened alumina can be described by a second-degree polynomial in the volume fraction of zirconia, given by Equation 3. The observed variations are also in close agreement with the theory based on spherical particle inclusion. The dependence on elastic moduli of the volume fraction of inclusions, also follows a similar equation. Thus a good correlation can be expected between the velocity and modulus. A plot of elastic modulus against ultrasonic velocity (Fig. 7) confirms this. Similar observations in other ceramics like uranium dioxide-sintered iron compacts and silicon carbide [2, 5], have already been reported in the literature. The data points are fitted into a straight line

$$E = -86.04 + 0.046\ V_l \quad (21)$$

with a $Q$ value of 0.92. Thus longitudinal velocity alone can be used for monitoring Young's modulus of

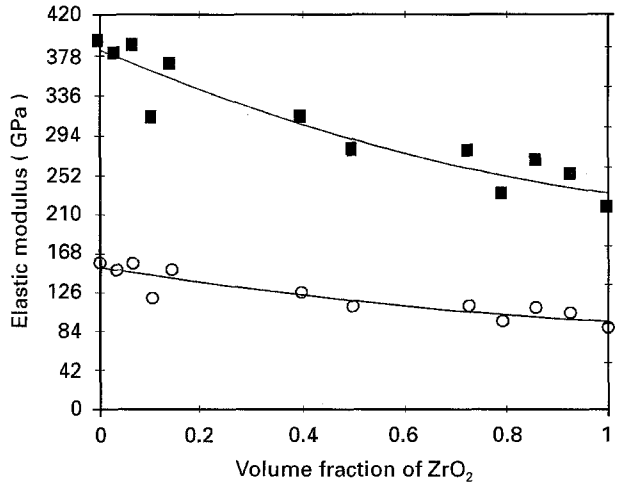


*Figure 5* Comparison of elastic moduli of sintered ZTA ceramics with the theoretically predicted values. (■) Young's, (○) shear moduli.

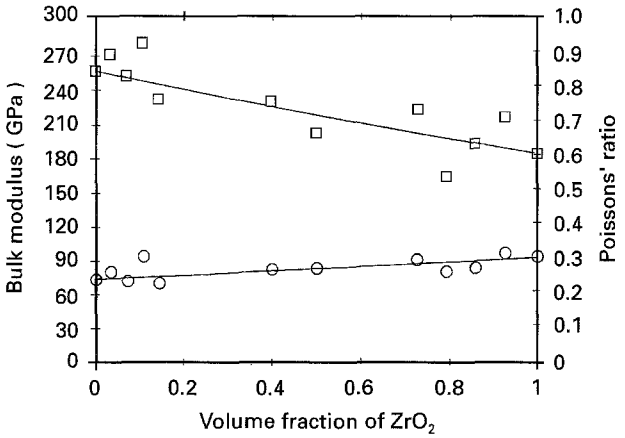


*Figure 6* Variation of (□) bulk modulus and (○) Poisson's ratio of sintered ZTA ceramics with volume fraction of $ZrO_2$. The theoretically predicted values.

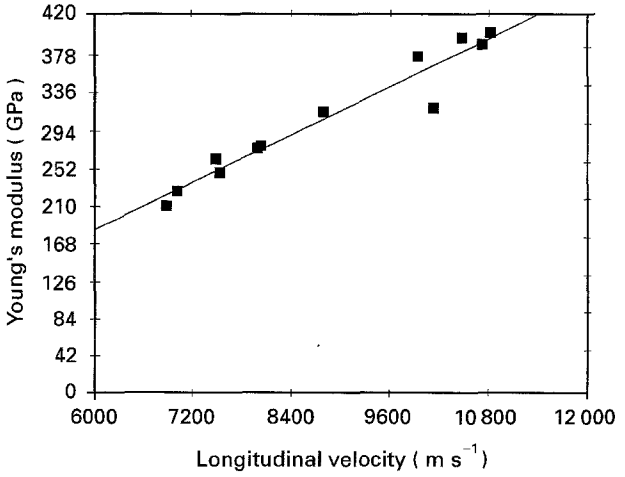


*Figure 7* Plot of Young's modulus against longitudinal ultrasonic velocity in ZTA ceramics showing a linear fit.

this ceramic as a routine check in a production plant. It may also be noted that Equation 10 can also be used to estimate the volume fraction of zirconia present from the longitudinal velocity measurements.

## 5. Conclusions

Ultrasonic velocity of zirconia-toughened alumina ceramics in the volume fraction of zirconia range from 0%–100% were generated. The measured velocity values were fitted into second-degree polynomials which would serve as calibration graphs for using ultrasonic velocity for monitoring the volume fraction of zirconia in this type of ceramic. The values were compared with the existing theory of elasticity models based on spherical inclusions. The observed variation with volume fraction of inclusion agreed well with the theory. The micrographs also confirm the spherical nature of the inclusion phase.

The elastic moduli were evaluated from the ultrasonic velocity data and, here again, they agreed with the values predicted by the elasticity theory quite well. Analysis of the data further shows that there is good linear correlation between longitudinal ultrasonic velocity and Young's modulus over the entire volume fraction range. The results show that ultrasonic velocity may be used as a predictor of elastic moduli for this two-phase ceramic.

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