

Ultrasonic Characterization

of Green and Sintered Ceramics:

II, Frequency Domain

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Nondestructive techniques, such as ultrasonics, have the capability of analyzing elastic as well as microstructural properties.¹ The conventional ultrasonic measurements conducted are those of velocity and attenuation. Ultrasonic velocity is considered as more of a macroscopic property and is mainly used for determination of elastic properties,²⁻⁴ although in many cases it may be used to determine microstructural changes. Ultrasonic velocity measurements have been used to characterize colloidally processed ceramics.⁵

This paper focuses on ultrasonic attenuation measurements of partially sintered, colloidally processed alumina using 5-30-MHz frequencies. Such measurements are very sensitive to the microstructure and can depend on a variety of parameters, e.g., the volume fraction and the nature of porosity, grains and second phases.

Theoretical treatment of scattering due to various microstructural features in a material is complex. A number of significant contributions have been made in the past toward understanding scattering from grains and from pores or inclusions of specific geometry. A notable contribution has been the

correlation between grain size distributions and attenuation measurements.¹ However, most of the treatments are devoted to independent scattering models of densified structures.

Understanding the attenuation of porous ceramics requires multiple scattering models in which scattering due to interaction between pores is considered. Although a number of significant contributions have been made in this direction,^{3,6} correlation with experimental data for higher volume fractions of porosity is difficult to obtain. This might be due to the complex nature of porosity in partially sintered structures, where pores are generally interconnected and can be of complex shapes, sizes and distributions.

Empirical approaches can provide some valuable insights in understanding the attenuation of partially sintered structures. An important observation made by Baaklini *et al.*⁷ is the relative sensitivity of attenuation measurements to density, pore shape and size of hot-pressed silicon nitride.

A continuous evaluation of attenuation measurements at various stages of densification might provide significant information about the state of microstructural evolution. To accomplish this, in the present study, relative attenuation measurements have been conducted on slip-cast alumina samples at various stages of densification.

Ultrasonic Attenuation

In this work, the relative attenuation was measured by a different technique than those described previously by other authors^{8,9} for frequencies ranging from 5 to 30 MHz. Broadband λ transducers,^{10,11} with clear-fused quartz (CFQ) as a reference, were used in the current study. This technique was successfully used by Bhardwaj *et al.*¹² for the analysis of superconductors.

In this technique, the test material is excited with special transducers (λ series) which have a very broad frequency spectrum analogous to the white spectra in optical spectroscopy. Thus, the incident frequency response selection of the material can be evaluated over a relatively large range of frequencies. This method uses a reference ultrasonic spectrum obtained by applying wideband transducers in direct transmission mode from a 1.0-cm-thick optically flat and polished CFQ.

Once the reference spectrum has been generated, similar spectra are obtained from test materials by sandwiching them between the transmitting and receiving transducers. Assuming that the reference spectrum corresponds to input frequency components, subtraction of the spectra of the test material from the reference spectrum, after normalizing for thickness, establishes the relative frequency dependence of ultrasonic attenuation as a function of

R&D Reviews are peer-reviewed articles of original research in engineering development and manufacturing practices from private, government and university laboratories around the world.

Manuscript No. 195028. Received December 17, 1992; approved October 7, 1993.

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various material characteristics.

It is important that, once the reference spectrum from the CFQ has been established, the pulser, amplifier or fast Fourier transform (FFT) mechanism settings are not altered during data acquisition.

Samples for ultrasonic analysis were sufficiently smooth and flat for efficient ultrasound transmission. A FFT was done to display the signal in the form of its frequency components. The ultrasonic attenuation was measured by sandwiching the top and bottom surfaces of the cylindrical samples between the transducers, so that the direction of ultrasound propagation was along the central longitudinal axis of the cylinder.

Despite the high attenuation of the partially sintered samples, a frequency range from 5 to 30 MHz was successfully used in this study. Although duplicate measurements were not conducted, an experimental error of about ± 2 dB/cm in attenuation measurements was expected but not conclusively determined.

Results and Discussion

The frequency spectrum of a 1-cm-thick CFQ is shown in Fig. 1 with a typical spectrum of a partially

sintered sample. After various samples were normalized for the thickness, their relative attenuation was obtained (in dB/cm) by subtracting from the frequency components of the reference CFQ. Relative attenuation was measured for dispersed (green density of 62.9%) and partially flocculated samples (53.9%) as a function of frequency.

The relative attenuations of the dispersed samples are, in general, lower than the partially flocculated samples, especially at lower densities (as shown in Figs. 2 and 3). At higher densities, the differences are not so apparent, perhaps because of the lower frequency range used in this study. The lower frequencies, however, seem to be sensitive to samples with lower densities.

Baakli *et al.*⁷ reported a 150% increase in the mean attenuation coefficient for a 1.97% decrease in density for samples of silicon nitride. However, they were able to work with extremely high frequencies (60–160 MHz) since their samples had high densities (90–98%). Since the densities in the present study were much lower (60–90%), a lower frequency range (5–30 MHz) was used. Assuming that the longitudinal ultrasonic

velocities for their silicon nitride samples ranged from 11,000 to 12,000 m/s, the wavelengths they used ranged from ~ 70 to 200 μm .

The theory of ultrasonic grain scattering for dense materials is generally studied in three different regions in which the frequency and grain-size dependencies of the attenuation is different. Depending on the value of λ/D (where λ is the wavelength and D the average diameter of the grains or dominant scatterer), these are the Rayleigh, stochastic and diffusive regions. The theory of scattering in these regions for polycrystalline materials has been considered by Papadakis.¹ The region of interaction in the work of Baakli *et al.*⁷ corresponds to the Rayleigh region ($\lambda > 2\pi D$), since the dominant scatterer is the small amount of pores (having median diameters from 2 to 4 μm) in their samples.

However, for the partially sintered alumina ceramics in the present work, the situation is different. Multiple Rayleigh scattering is expected to be the dominant scattering mechanism considering the proximity of the pores, if one assumes the pores (about 0.5 μm) to be the dominant scatterer. The contribution from grain boundaries

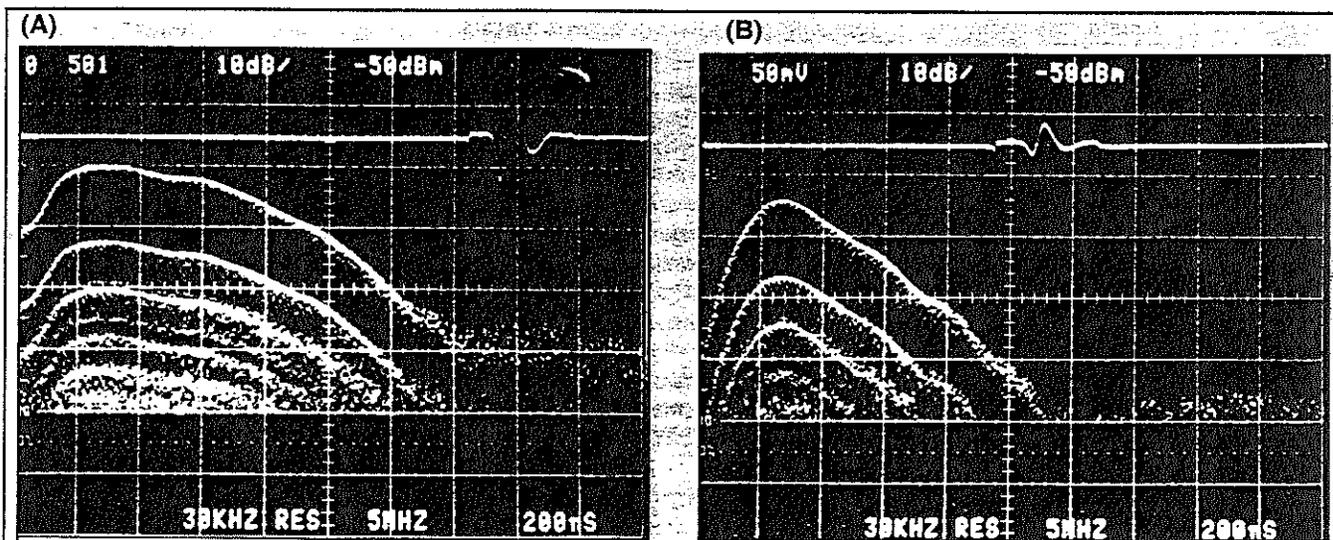


Fig. 1: Time and frequency domain analysis of (A) CFQ and (B) partially sintered alumina.

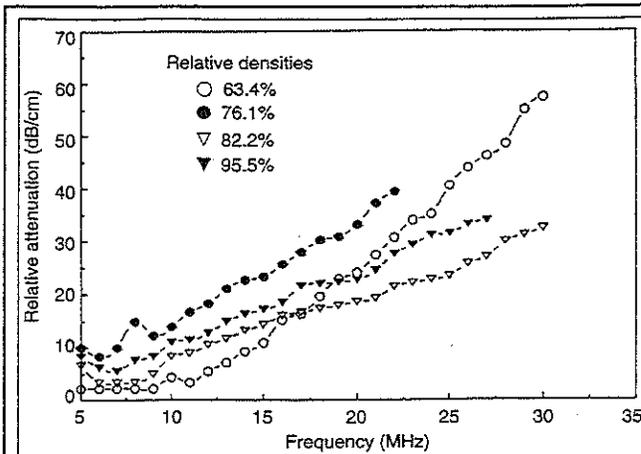


Fig. 2. Relative attenuation as a function of frequency for samples having green densities of 62.9%.

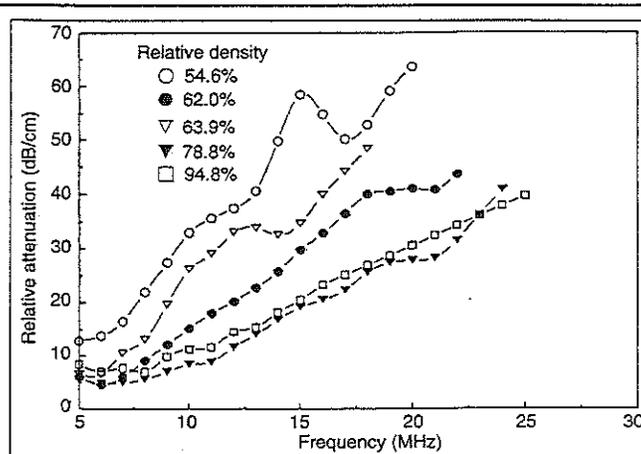


Fig. 3. Relative attenuation as a function of frequency for samples having green densities of 53.9%.

(mean grain size from 1 to 5 μm for polycrystalline alumina) toward scattering for the frequencies used in the present work is expected to be negligible compared to that from the pores, especially for the partially sintered samples. Besides the difference in scattering mechanisms, the relative attenuation and not the absolute attenuation¹³ was measured in this work.

It is expected that, for the partially sintered samples, besides the total volume fraction of the porosity, the tortuosity of the pore-grain boundary interface, size, shape, connectivity and distribution of pores also may have some contribution to attenuation. It has been shown by Evans *et al.*¹⁴ that the attenuation is affected mostly by the dominant scatterers present in the ceramic.

At low densities, because of the higher pore sizes and the randomness in the microstructure, the relative attenuation of the partially flocculated samples is expected to be higher, as is observed here. The scenario at higher densities does not appear to be clear from this investigation, although it is expected that the partially flocculated samples have higher attenuations, even at >95% relative densities. The scattering mechanism here is expected to be controlled by the grain-size distribution and the

relative sizes of the remaining pores.

Conclusions

The relative attenuation measurements (5–30 MHz) conducted in this work were sensitive to the microstructures of the partially sintered samples at lower densities. The partially flocculated samples showed higher attenuations than the dispersed samples. Thus, the effect of the prior history and processing of these samples was reflected in the attenuation measurements at an early stage in the sintering process.

It is currently difficult to account for the attenuation behavior for the complex range of microstructures encountered in this work, although a few qualitative trends are expected. Clearly, more detailed investigations are needed, because different types of scattering mechanisms seem to be operating for various microstructures, depending on the particular frequency. Hence, the relation expected between attenuation and porosity may not be easily evident at this time. It is expected that high-frequency attenuation measurements are more sensitive to microstructural changes in such samples. Proper quantification of the microstructure and detailed attenuation measurements during various stages of sintering are neces-

sary to properly account for the observed attenuation behavior. Use of various stereological parameters need to be explored as a means of quantifying the microstructure of partially sintered structures.¹⁶ For example, the gross tangent count,¹⁷ which is a measure of randomness in the microstructure, might be used to explain attenuation behavior of partially sintered structures. The use of attenuation measurements of partially sintered ceramics as a quality-control tool appears promising. ■

Acknowledgment

N. Kulkarni and B. M. Moudgil acknowledge the National Science Foundation (MSS 8821815) for the financial support of this work.

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Editor's Note: Part I of this two-part series was published in the June 1994 issue of Ceramic Bulletin (pp. 146–53). In Part I, the authors reported that ultrasonic velocities of colloidal processed alumina—measured using dry transducers—were affected by density and particle packing. Experimental details and methods of characterization were presented.

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