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PROCESS MONITORING USING OPTICAL ULTRASONIC WAVE DETECTION

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ABSTRACT

Optical ultrasonic wave detection techniques are being developed for process monitoring. An important limitation on optical techniques is that the material surface, in materials processing applications, is usually not a specular reflector and in many cases is totally diffusely reflecting. This severely degrades the light collected by the detection optics, greatly reducing the intensity and randomly scattering the phase of the reflected light. A confocal Fabry-Perot interferometer, which is sensitive to the Doppler frequency shift resulting from the surface motion and not to the phase of the collected light, is well suited to detecting ultrasonic waves in diffusely reflecting materials. This paper describes the application of this detector to the real-time monitoring of the sintering of ceramic materials.

INTRODUCTION

Certain microstructural features of materials, such as grain size in metals, porosity in ceramics, and structural phase compositions, are important for determining mechanical properties. Many of these microstructural features have been characterized by ultrasonic wave propagation measurements, such as wave velocity and attenuation. Real-time monitoring of ultrasonic wave propagation during the processing stage would be valuable for following the evolution of these features. This paper describes the application of laser ultrasonic techniques to the monitoring of ceramic sintering. Prior to this work, ultrasonic wave measurements of the sintering of ceramics have been made only through direct contact with the material with a buffer rod [1,2]. Recently, several advances have been made using lasers for both generation and detection of ultrasonic waves in a totally noncontacting manner for material microstructure evaluation [3-5]. Application of laser ultrasonic techniques now opens the possibility for real-time monitoring of materials in very hostile environments as are encountered during processing [6].

Zinc oxide was chosen as the ceramic material to be studied as it readily sinters between 850 and 950°C. Laser ultrasonic measurements are presented for samples of zinc oxide with rough and/or optically diffusely reflecting surfaces, including in situ measurements taken during sintering.

EXPERIMENTAL MEASUREMENTS

Figure 1 is a schematic of the system for laser ultrasonic measurement. A pulsed Nd-YAG laser with a 10 ns pulse width and pulse energies of up to 300 mJ was used as the ultrasonic source. The laser beam was focused down to a diameter of approximately 3 to 4 mm and produced some ablation of material from the surface, which was evident from discoloration. The initial powder compacts, green state samples, were particularly soft with roughly the consistency of chalk. These materials were ablated significantly by the source laser at the energy levels required for adequate signal to noise ratio. Therefore, the ultrasonic response of the green state samples was recorded with a minimum of signal averaging, usually 10 pulses. Surface ablation was significantly reduced after the samples sintered to greater than 50% theoretical density.

A confocal Fabry-Perot interferometer, modeled after that described in reference [7], was used to detect the ultrasonic waves on the surface of the samples in a through transmission configuration. The detector is sensitive to the Doppler shift of light reflected from the sample surface, which is moving due to the ultrasonic wave. An argon ion continuous laser (maximum output of about 1.5 W in the green line) was used to illuminate the sample surface. A small portion of the beam was diverted for electronic stabilization of the Fabry-Perot cavity length with respect to the laser frequency. The electronic stabilization corrected for the laser drift and low frequency instability due to ambient vibrations of the apparatus and allowed the entire apparatus to be mounted on a fixed table without additional isolation. This unit is sensitive only to the frequency of the reflected light and not its phase, which allows it to collect light from a relatively large area (about 1 mm^2) on the surface. This capability gives it the sensitivity needed for detection from rough and/or optically diffuse material surfaces, as encountered in ceramic materials in general and presintered materials in particular.

Figure 2 shows the ultrasonic waveforms detected from polished and unpolished opaque samples of silicon nitride. The pulsed laser was the source for the ultrasonic waves on the opposite side of the sample. The

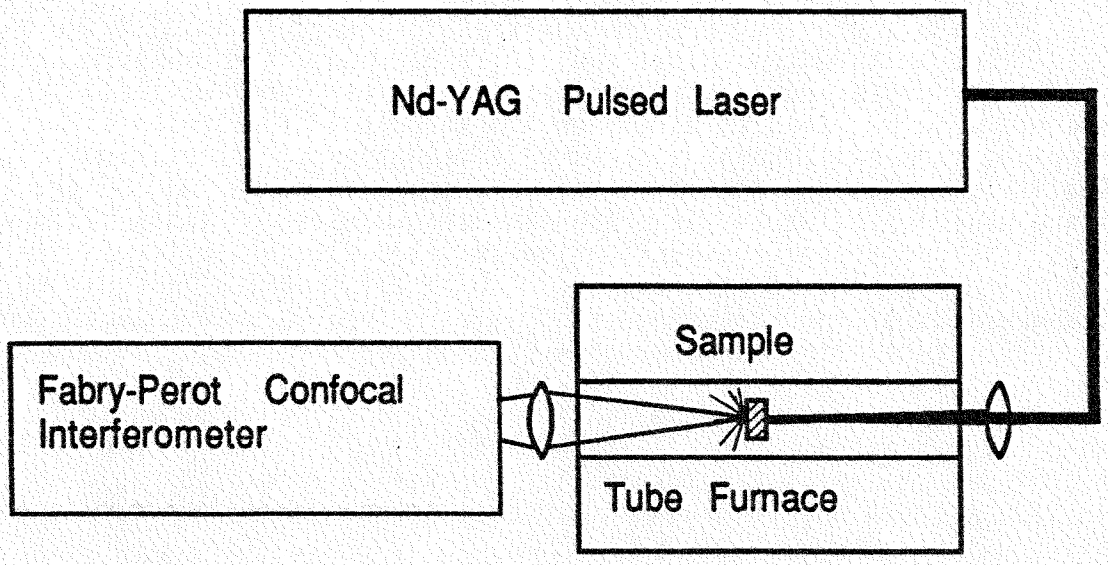

Fig. 1. Block diagram of the laser ultrasonic experiment. The pulsed laser has a pulse width of 10 ns at 1.06 μm. The interferometer u)es an argon ion laser with 1.5 W in the green line, 0.514 μm.

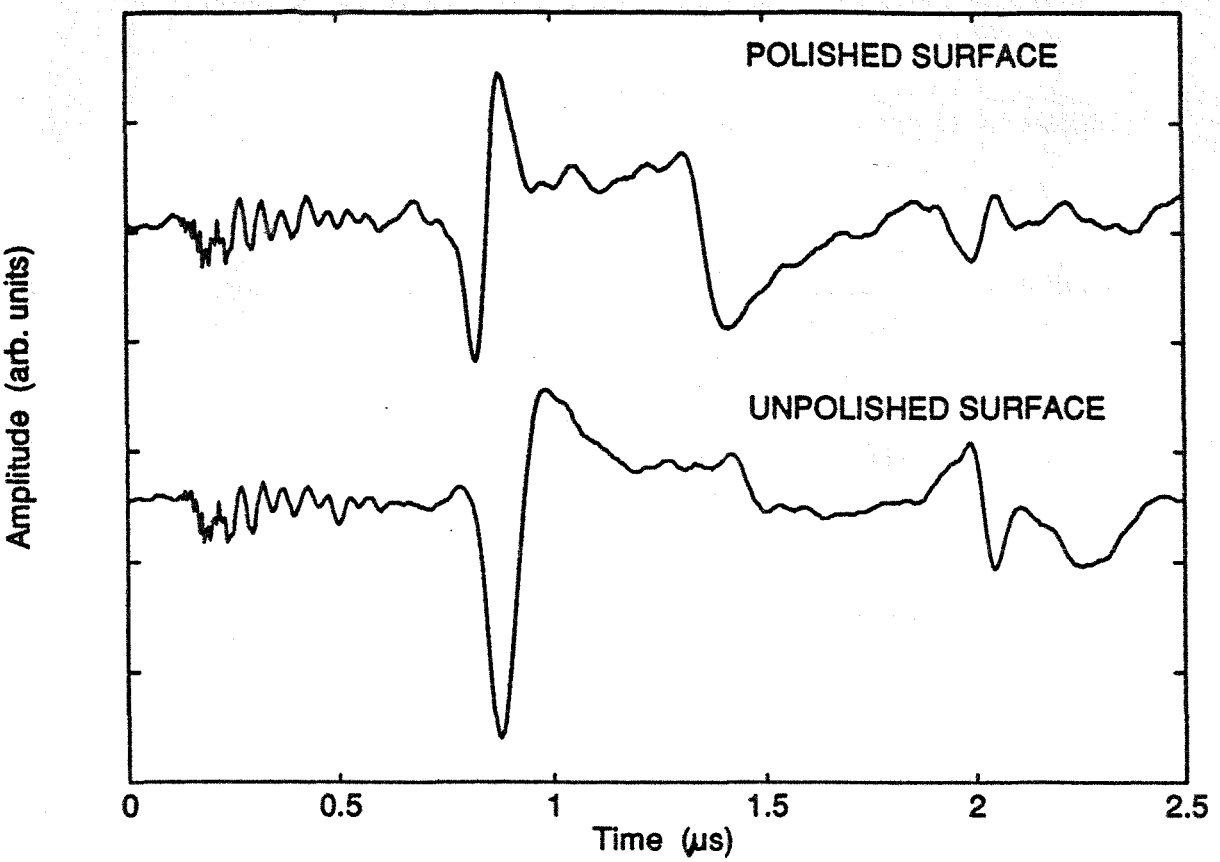

Fig. 2. Comparison of laser generated ultrasonic waveforms recorded with the Fabry-Perot interferometer for two samples of silicon nitride. The 6.1 mm thick samples were identical for surface finish. The unpolished sample was coarse ground with 120 grit paper and the polished sample had a mirror-like surface. Detector laser powers were 1.6 mW (polished) and 50 mW (unpolished), respectively.

figure shows that very similar waveforms were recorded from the unpolished and polished samples with essentially the same signal to noise ratio. This was achieved by increasing the argon ion laser power from 1.6 mW for the polished sample to 50 mW for the unpolished sample; much of the lack of specular reflection due to the rough surface can be compensated for by increasing the detection laser power. No surface damage resulted from using the detection laser at these powers. The difference between the polished and unpolished results, evident in Figure 2, is under further investigation. The zinc oxide samples used for the sintering study exhibited nearly completely diffuse reflection from their surfaces, even after sintering.

POST-SINTERING RESULTS

Several samples of zinc oxide were prepared in the green state by compressing the powder into preforms about 25 mm in diameter by 10 mm thick with relative densities (compared to theoretical density) of about 47%. Samples were then sintered. The temperatures used ranged from 800 to 960°C and the times from 2 min to 2 h; sintering parameters which would produce a set of partially sintered samples with relative densities ranging from 52 to 97% were chosen. Longitudinal ultrasonic wave velocities were recorded for this sample set to provide basic information on the effect of sintering. Measurements were taken with a 5 MHz contact piezoelectric transducer, using a vacuum coupled polymer film (similar to Robert's technique [8]) with gel couplant, and the laser ultrasonic setup

as shown in figure 1. Both measurement methods agreed to within 0.5% on the resultant longitudinal wave velocities; the major error came from the thickness measurement.

Figure 3 shows several of the ultrasonic waveforms recorded with the laser technique for samples of various relative densities. Detection laser powers of about 150 mW were used for these measurements. The figure shows a well developed ultrasonic waveform for all samples that have been partially sintered (>52%). The major change in the ultrasonic waveforms among the various samples is the significantly decreased time of flight as the sample's density is increased, a 25% reduction in thickness was observed between the 47% dense sample and the 97% dense sample. As the relative density increased in the samples, the wave velocity increased significantly and the attenuation decreased.

The measured longitudinal wave velocities for the sample set, obtained with both the piezoelectric and laser techniques, are shown in Figure 4. The velocities are low for the green state material, around 0.7 mm/μs, but rise abruptly after a small amount of sintering. This rapid rise in velocity, as depicted by the 52% samples, is probably due to joining between the powder particles in the initial sintering. This results in a large change in elastic constant for the sample but very little densification. Further sintering produces a roughly linear change in velocity with relative density up to values approaching those of the bulk material as full densification is achieved. This velocity behavior provides a very straightforward method for monitoring the sintering process and densification in particular.

REAL-TIME SINTERING EXPERIMENTS

Laser ultrasonic measurements have also been made on zinc oxide samples during the sintering process. Figure 5 shows the waveforms recorded while a sample was heated to 850°C. In the figure, trace (a) was taken on the sample at room temperature before insertion into the furnace. Several minutes were taken to insert the sample into the tube furnace, and trace (b) was recorded upon completion of the insertion. At 850°C the material sinters to nearly theoretical density in about 2 h.

At the high temperature in the furnace the sample reflectivity was significantly lower than at room temperature. The detection laser power was increased to 900 mW in order to record the waveforms of figure 5. The waveforms show a rapid decrease in the ultrasonic time of flight for the first 10 min after insertion. This is due to the bonding between particles in the powder, the changing sample thickness, and the increasing densification taking place. Thus far no independent measurement of the sample thickness has been made. In the near future thickness measurement capabilities will be added so that the velocity can be determined and densification recorded directly from the ultrasonic waveforms. Figure 5 shows that the laser ultrasonic technique has sufficient signal to noise ratio to monitor the sintering of zinc oxide as a function of time.

CONCLUSIONS

Laser ultrasonic techniques have been successfully applied to the sintering of zinc oxide. The Fabry-Perot interferometer was used, both during and after sintering, to record ultrasonic surface motions of samples with relative densities of 47 to 97% of theoretical. For partially sintered samples, the longitudinal wave velocity was essentially linearly dependent on the relative density. We conclude that the

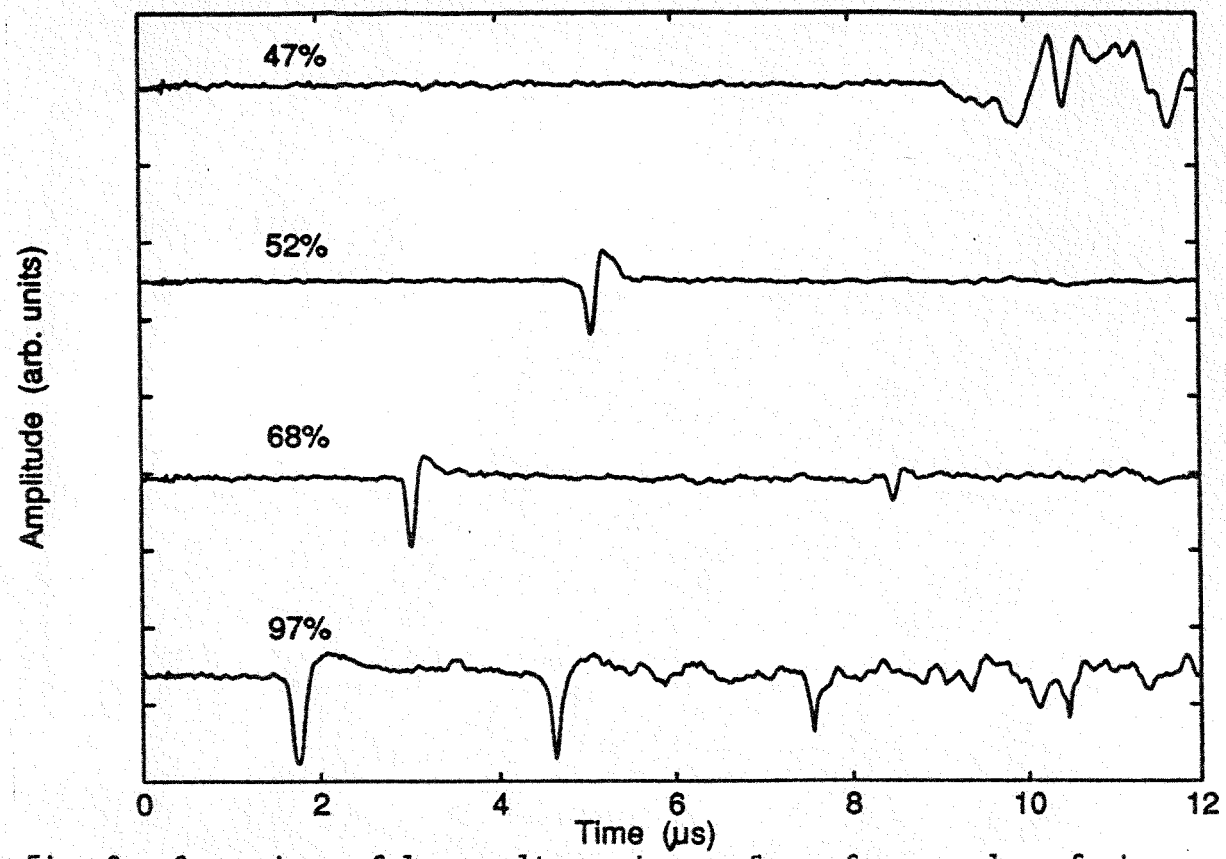

Fig. 3. Comparison of laser ultrasonic waveforms from samples of zinc oxide presintered to different relative densities. The source is the pulse laser, with energies of around 100 mJ/pulse, and the detector is the Fabry-Perot interferometer with 150 mW for the argon ion laser.

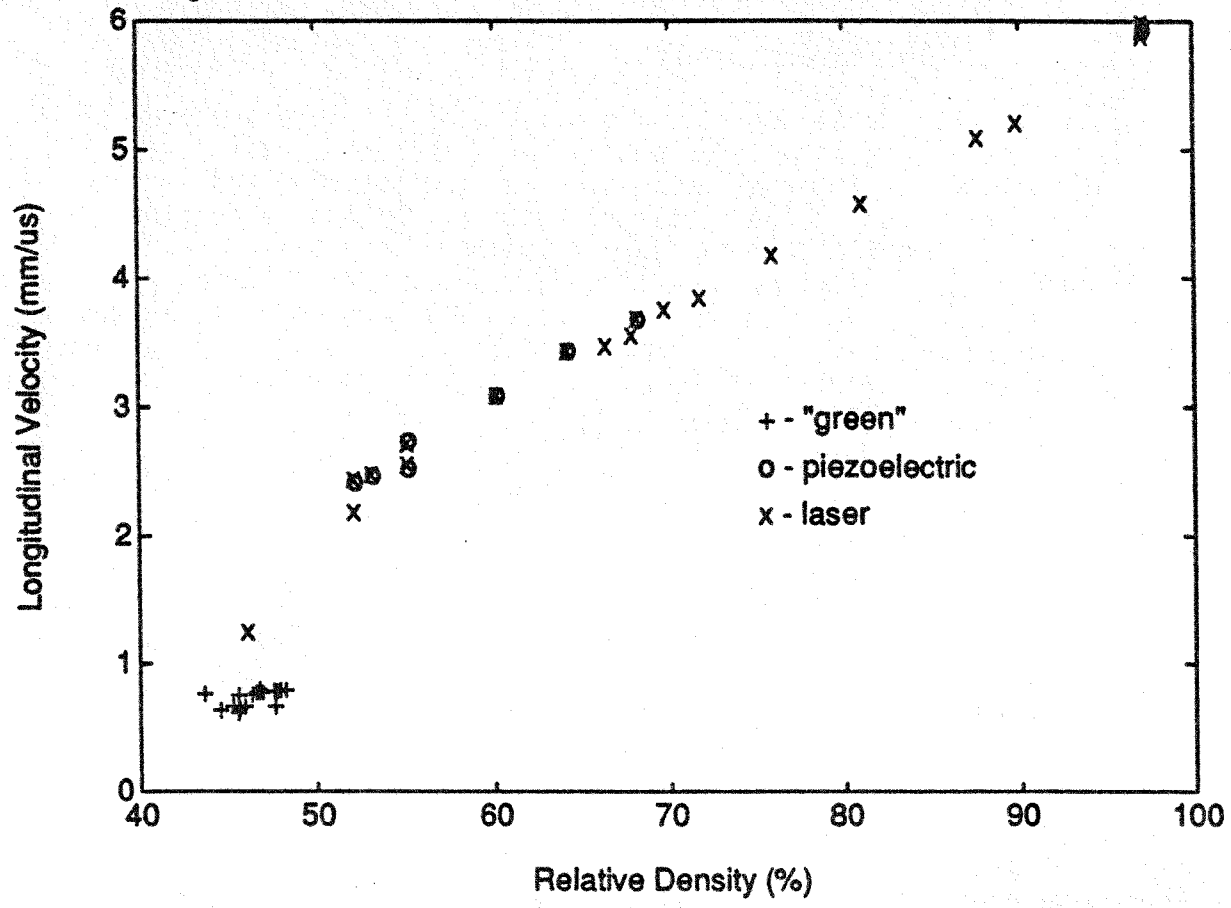

Fig. 4. Longitudinal wave velocities of the presintered zinc oxide sample set measured with both the piezoelectric and laser techniques.

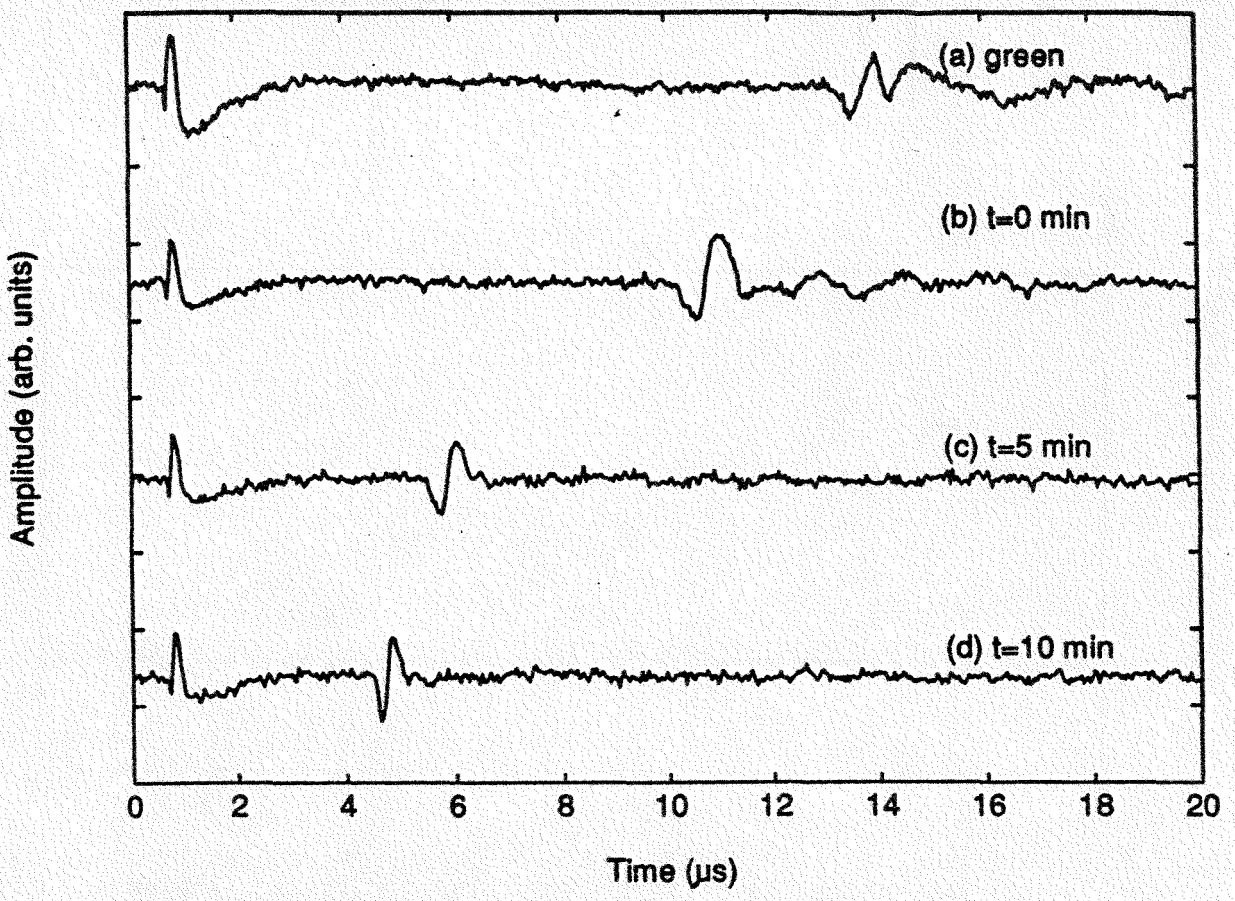

Fig. 5. Real-time laser ultrasonic measurements in the through transmission mode for zinc oxide. The measurements were taken while the sample was heating in a tube furnace at 900°C. The firing of the pulsed laser is marked by the optical pulse near the origin for each A-scan.

ultrasonic velocity provides a direct measure of the densification of the material. Several different factors in the sintering process are still unknown, such as the internal temperature of the material and its net shape (thickness) during the sintering process. With a method for measuring sample shrinkage, the laser ultrasonic measurement of densification will allow the direct monitoring of the sintering of ceramic materials as a function of both time and temperature, which will provide valuable information for the optimization of this material fabrication process. This technique should also be applicable to other material processing environments, such as annealing in metals.

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REFERENCES

1. J. H. Gieske and H. M. Frost, "Sound Velocity Measurements in Green-Body Ceramics as a Function of Sintering Temperatures," Reviews of Quantitative Nondestructive Evaluation, 8B, D. O. Thompson and D. C. Chimenti (eds.), Plenum Press, New York, 1709 (1989).

2. F. Duvant, J. du Mouza, M. Arnould, and R. Struillou, "Ultrasonic Control of Ceramic Products Cracking During Their Making Between 20 and 1400°C," Proceedings of the Second International Symposium of Ceramic Materials and Components for Engines, Verlag Deutsche Keramische Gesellschaft, Bad Honnef, West Germany, 849 (1986).

3. C. B. Scruby, R. L. Smith, and B. C. Moss, "Microstructural Monitoring by Laser-Ultrasonic Attenuation and Forward Scattering," NDT International, 19, 307 (1986).

4. L. Piche, B. Champagne, and J.-P. Monchalin, "Laser Ultrasonics Measurements of Elastic Constants of Composites," Materials Evaluation, 45, 74 (1987).

5. K. L. Telschow, "Microstructure Characterization with a Pulsed Laser Ultrasonic Source," Reviews of Quantitative Nondestructive Evaluation, 7B, D. O. Thompson and D. C. Chimenti (eds.), Plenum Press, New York, 1211 (1988).

6. R. J. Dewhurst, C. Edwards, A. D. W. McKie, and S. B. Palmer, "A Remote Laser System for Ultrasonic Velocity Measurement at High Temperatures," J. Appl. Phys., 63 (4), 1225 (1988).

7. J.-P. Monchalin and R. Heron, "Laser Ultrasonic Generation and Detection with a Confocal Fabry-Perot Interferometer," Materials Evaluation, 44, 1231 (1986).

8. R. A. Roberts, "Ultrasonic NDE of Green-State Ceramics by Focused Through-Transmission, Reviews of Quantitative Nondestructive Evaluation, 6B, D. O. Thompson and D. C. Chimenti (eds.), Plenum Press, New York, 1443 (1987).