NONDESTRUCTIVE CHARACTERIZATION OF GREEN AND SINTERED CERAMICS

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ABSTRACT

It is well-known that NonDestructive Characterization (NDC) can enhance our knowledge of materials and processes, reduce cost of production and energy usage, and establish criteria of materials reliability in our increasingly complex world.

However, despite the long history of ultrasonic testing, this subject is still new in the field of materials development and manufacture. For example, if ultrasound can be coupled to materials **without altering their original features**, then it is relatively simple to define their densities, mechanical properties, microstructure, defects, interfaces of multi-layered materials, anisotropy, etc. - all the way from green to sintered stages. In this paper we describe the significance and the principle of materials-suitable *modus operandi* of ultrasonic NDC for green and sintered ceramics.

INTRODUCTION

Investigation of transmitted ultrasound through a material can provide valuable information about it (1). Triggered by the diverse and intense industrial demands - process and quality control, failure prevention, safety assurance, *in-situ* diagnostics, efficient and cost effective use of energy and raw materials, etc. - widespread applications and new trends are being developed in NDC of materials (2,3,4,5). Properly transmitted ultrasound in a material - whether under RTP, or under extreme physico-chemical environment - and its correlation with material parameters can greatly assist manufacturers and users in materials quality and safety endeavors. Table 1 summarizes the functional significance of ultrasonic NDC.

However, for reliable uses of ultrasound and subsequent data interpretation, we must first consider some basic issues. They are important not only from the standpoint of adhering to the strict theme of the test material's **"nondestruction and maintenance of its original features,"** but these issues also recognize the physico-chemical diversity of materials at various stages of their manufacture.

TRANSDUCER Coupling and its Significance for Ceramics NDC: How a transducer is coupled to a test material is extremely important for the NDC of ceramics and other powder- and fiber-based materials, which are characterized by enormous compositional and mictoructural diversities compared to most primary metals. Without the consideration of limitations, the extension of *metals-type conventional NDT* practice to ceramics is not suitable (6). One of the major limitations of conventional NDT is the use of liquid couplants at the transducer and test material interface. For obvious reasons, the use of liquid couplants for the analysis of green, relatively

Table 1. Significance of ultrasonic NDC, including measurement categories and corresponding measured parameters.

MEASUREMENT CATEGORY	MEASURED PARAMETERS	INFORMATION REVEALED & APPLICATIONS
TIME DOMAIN	Velocities of longitudinal, shear, and surface waves	Density, porosity, defect detection, elastic and mechanical properties, interface analysis, anisotropy, etc.
FREQUENCY DOMAIN	Frequency dependence of ultrasonic attenuation (Ultrasonic Spectroscopy)	Microstructure: grain size and grain boundary relationships, porosity, etc.
IMAGE DOMAIN	Time of Flight, velocity, and attenuation mapping	Surface and internal imaging of defects, microstructure, density, velocity, etc.

porous, and liquid and environmentally-sensitive materials, will yield erroneous and unreliable ultrasonic observations, and destroy the test material during the process of NDC.

In order to circumvent this problem, about 10 years ago we developed a simple, but novel dry coupling technique. At the heart of this technique is the replacement of *"hard and high acoustic impedance protective face"* (typical of conventional wet-coupling transducers) by an *"acoustically transparent solid compliant transitional layer"* in front of the active piezoelectric element, Fig. 1. Characteristics of this transitional layer, which is an integral part of the direct contact, and

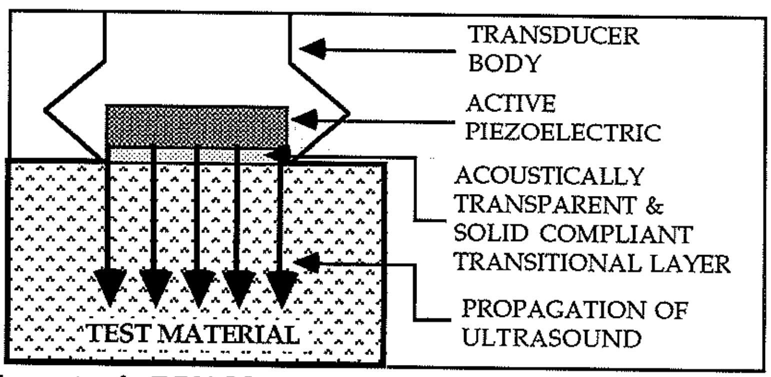

Fig. 1. Critical elements of a DRY COUPLING transducer. The acoustically transparent and solid compliant transitional layer efficiently transfers ultrasound from the active piezoelectric into the test material without liquid coupling - a limitation of conventional ultrasonic testing.

replaceable part for delayed contact devices, have resulted in the successful development of dry coupling longitudinal and shear wave transducers from <100KHz to >25MHz. Pulse widths of such devices can be controlled from a couple of wavelengths (for high resolution) to several wavelengths (for high sensitivity) for various applications. However, it is also important to note that in conjunction with transducers characterized by extremely short pulse ($\lambda/2$) and near "white" frequency spectra, the dry coupling mechanism has also been used for microstructure characterization of ceramic superconductors and other microstructurally heterogeneous materials (7,8). The feasibility and reliability of green and sintered ceramics NDC are shown by the application of this basic and simple development in ultrasound.

PROCEDURE

A computer-based (IBM PC Compatible) pulser/receiver system (NUSON NDC 5010) was used to excite and amplify ultrasonic signals from dry coupling transducers in direct transmission mode, Fig. 2. Typical steps for TOF (time of flight) and velocity measurement are shown in Fig. 3. The dry coupling transducers were hand held with "finger pressure." The significance of this

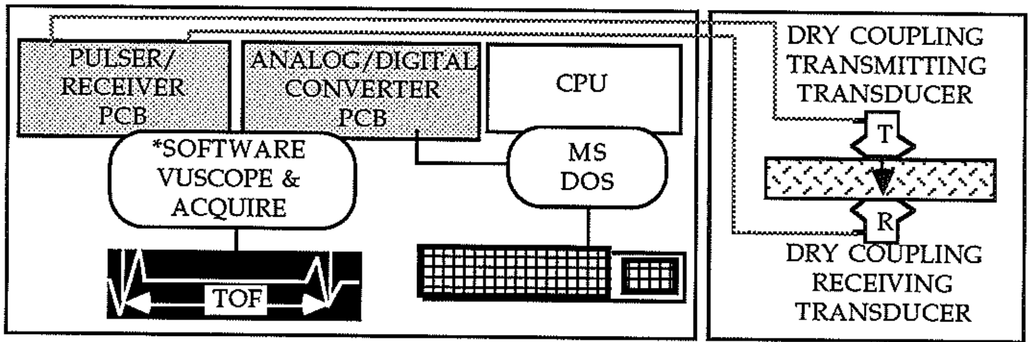

Fig. 2. **Schematic layout of NDC 5010 ultrasonic system used in this investigation.** ***Software VUSCOPE converts the computer monitor into a functional oscilloscope. ACQUIRE is used for time and frequency domain analysis.***

procedure is to accurately establish the "zero" reference point. This is accomplished by placing the left hand cursor (on a computer monitor or on an oscilloscope) at the trailing edge of the transmitted signal, corresponding to transit time through transducer matching layers when they are in contact with each other, Fig. 3, middle portion. By using this technique, accuracy and repeatability of the data to >99.5% have been consistently observed. In order to speed up the process of velocity determination, transducers can also be mounted on electronic digital callipers. This mechanism feeds the material thickness into the computer for direct velocity readout.

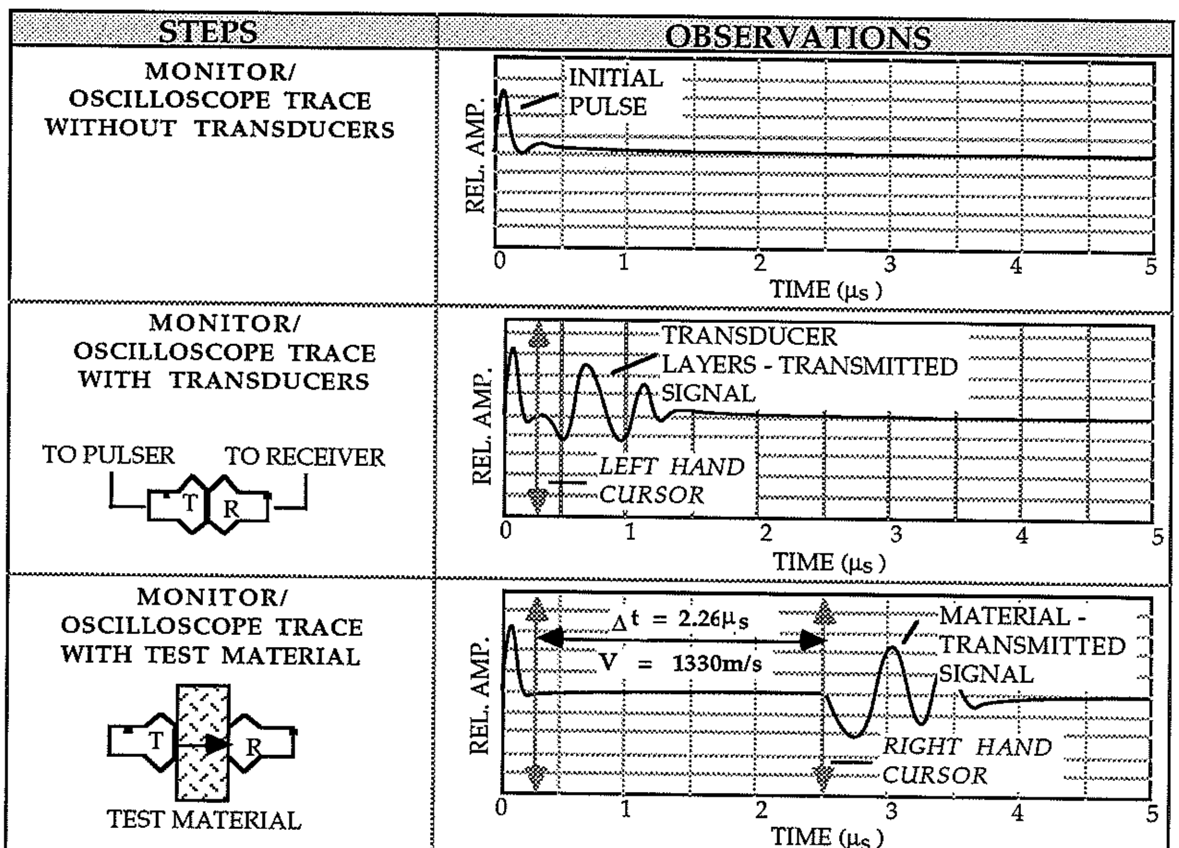

Fig. 3. **Steps and corresponding observations for time of flight and velocity measurement by DRY COUPLING technique. Example shown is for a 3mm green Al_2O_3 sample analyzed by 2MHz transducers. This technique is also suitable for shear wave velocity measurement.**

Besides establishing density-velocity relationships for green and sintered ceramics, longitudinal (V_l) and shear (V_t) wave velocities were also used to determine their elastic properties by using the following well-known relations:

E (Young's Modulus) = $V_l^2 \cdot \rho \cdot (1+\sigma) \cdot (1-2\sigma)/(1-\sigma)$ (1)

G (Shear Modulus) = $V_t^2 \cdot \rho$ (2)

σ (Poisson's Ratio) = $(1 - 2b^2)/(2-2b^2)$, where, $b = V_t/V_l$ (3)

K (Bulk Modulus) = $E/[3(1-2\sigma)]$ (4)

OBSERVATIONS AND RESULTS

GREEN Ceramics: Six batches (10 samples per batch) of sub-micron Al_2O_3 were compressed with PVA (poly vinyl alcohol) at varying compaction pressures to yield different densities.* Each sample, 25mm diameter and 3mm thick, was analyzed at five points by 6mm active area diameter and 2MHz longitudinal and shear wave dry coupling transducers. By assuming the average of five velocities to be the reference for each sample, they were plotted against their respective densities, Fig. 4. A similar relationship for Young's modulus is shown in Fig. 5.

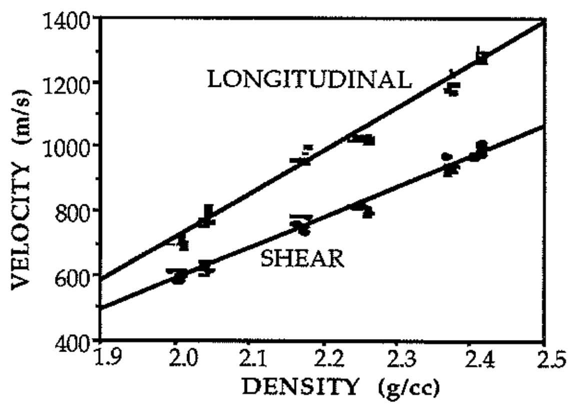

Fig. 4. Green Al_2O_3 - density vs. longitudinal and shear wave velocities relationships.

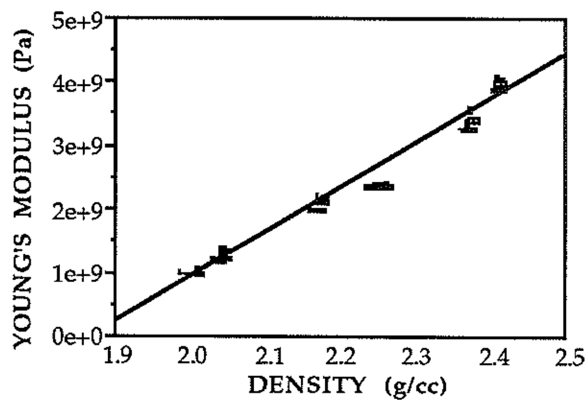

Fig. 5. Green Al_2O_3 - determination of Young's Modulus from Fig. 4 data.

By using relationships in Figures 4 and 5 for Al_2O_3 and similar ones for other materials, green components - steatite, cordierite, ZrO_2, TiO_2, WC, ferrites, powder metals, etc. - can be easily characterized for discrete point density variations as well as for internal cracks and voids.

This technique has been suitably used for density and defects characterization of green components such as tapes (~0.1mm) and large refractories (>1m). It is however, important to note that due to high frequency-dependence of ultrasonic attenuation, coarse grained and very large shape green components require relatively lower frequencies and higher energy ultrasonic systems. It is generally arduous, if not impossible, to analyze small cylindrical green shapes from the circumferential directions. Table 2 provides applicable frequency ranges and detectability of internal defects for various green materials. These relations were developed in our laboratory.

SINTERED Ceramics: In order to exhibit the applicability and reliability of the dry coupling technique for sintered ceramics NDC, two extreme examples of microstructure were chosen: 1. Dense Al_2O_3,** and 2. "Ultra" porous rigid ceramic preforms.*** Fig. 6 shows a relationship between density and Young's modulus of fully sintered Al_2O_3 as a function of density, determined by the dry coupling technique. Similar data for Al_2O_3, obtained by standard 4-point bending method, is also shown for comparison purpose. Fig. 7 shows similar relationships for highly porous ceramic preforms. The destructive method data in Fig. 7 is after reference (10). While

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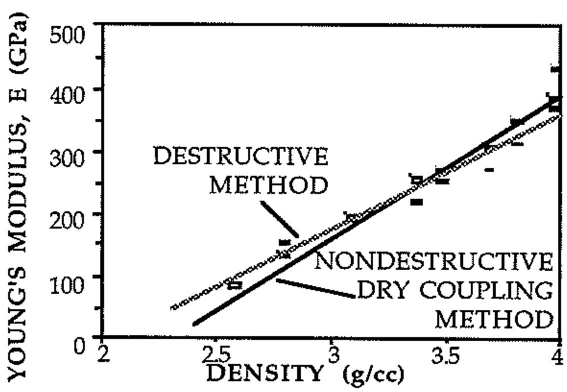
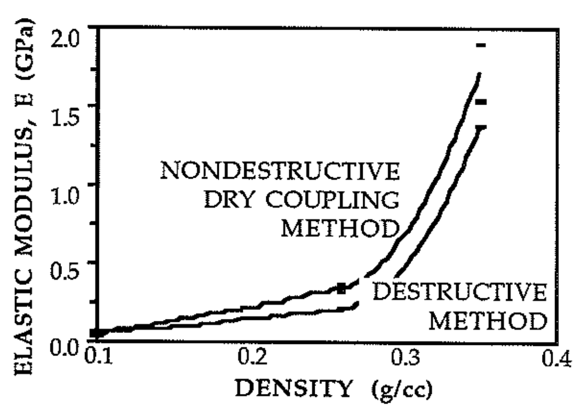
Table 2. A general description of applicable frequencies and defect detectability for the NDC of green materials.

GREEN MATERIAL	*FREQUENCY RANGE (MHz)	*MINIMUM DETECTABLE DEFECT ~(mm)
Fine grain -Dense (electronic & engineering ceramics, composites, etc.)	2-20	0.1
Medium grain - Dense to semi-porous (refractories, cermets, etc.)	0.5-2	>1.0
Coarse grain - Semi-porous to porous (refractories, concretes, cements, etc.)	<0.2-1	>5.0

**These are generalized statements provided for reference purpose only. For example, in some cases, while it may appear that relatively higher frequencies are usable, however, the actual frequencies transmitted through green materials may be much lower. When such observations occur, the detectability would be correspondingly reduced. Discussion on this subject is beyond the scope of this paper (9).*

examining relatively porous materials by the dry coupling technique, it is apparent that their "true" state is being characterized. Use of liquid couplants would, obviously, alter the original state of porous materials!





Comparison of destructively (4-point bending method) and nondestructively (DRY COUPLING ultrasonic method) determined elastic modulus of DENSE and POROUS ceramics.

Fig. 6. Sintered Al_2O_3 varying from ~70 to 100% theoretical density.

Fig. 7. "Ultra" porous ceramic preforms with 50 to >80% porosity.

CERAMIC Composites: Table 3 shows acoustic and elastic properties of single and multiphase SiC and Si_3N_4 determined by 20MHz and 10MHz longitudinal and shear wave dry coupling transducers, respectively. It is important to note that relatively subtle density variations in these materials are also reflected by their respective longitudinal and shear wave velocities, thereby demonstrating the sensitivity of this NDC technique.

FIBER-BASED Composites: Materials characterized by directional and heterogeneous properties, including porosity, can also be analyzed by the dry coupling technique. This is demonstrated by the examination of a 2D carbon-carbon (C-C) composite. This material is useful for high temperature and high strength applications and is highly anisotropic; thus, it exhibits two sets of properties, i.e., along and across the directions of carbon fiber orientation. The degree of anisotropicity is easily revealed by ultrasonic velocities in these directions as a function of density, Fig. 8. It is interesting to note that, in the direction across carbon fibers, this material requires relatively low 0.25 to 0.5MHz frequency due to high frequency-dependent attenuation. However, in the direction along fibers, higher 5MHz frequency was suitable due to relatively lesser frequency-dependent attenuation. This information is considered vital from the standpoint of processing and applications of C-C fiber composite.

Table 3. Ultrasonically determined properties of single and multi-phase SiC and Si_3N_4.

MATERIAL	DENSITY (g/cc)	VELOCITY (m/s) LONGITUDINAL	SHEAR	ELASTIC MODULI (GPa) E	G	K	POISSON'S RATIO σ
SiC - Single Phase	3.01	11,500	7,320	374	161	184	0.161
	3.09	11,800	7,530	406	175	198	0.159
	3.15	12,050	7,700	431	186	210	0.157
SiC - Composite*	2.37	7,480	4,640	121	51.1	64.8	0.186
	2.40	7,540	4,750	126	54.0	64.2	0.172
Si_3N_4 - Single Phase	3.05	11,050	6,225	299	118	215	0.268
Si_3N_4 - Composite*	2.30	6,390	4,010	86.6	36.9	44.3	0.174
	2.35	6,540	4,020	90.8	38.0	50.0	0.196

**Second phase, presumably amorphous.*

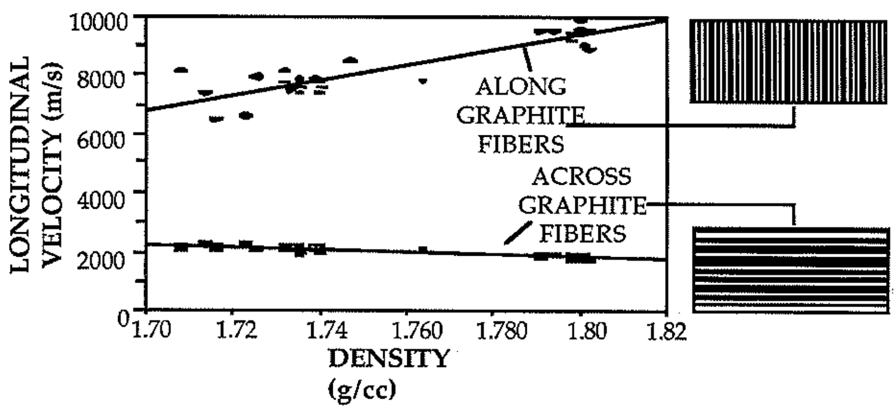


Fig. 8. Velocity relationship of 2D Carbon-Carbon composite as a function of fiber orientation, exhibiting anisotropic characteristics of this material. A similar trend was also observed for elastic modulus.

<u>OTHER Ceramics and Composites:</u> A number of conventional and advanced ceramic materials, varying in composition and microstructure, have been characterized by the dry coupling technique described in this paper. The diversity of acoustic and elastic properties of some selected materials is shown in Table 4.

CONCLUSIONS

In this paper we have described the simple, yet significant dry coupling ultrasonic technique for its relevance to the nondestructive characterization of green and sintered materials. In order to exhibit the feasibility and reliability of this technique including its significance, several examples have been provided of a wide range of conventional and modern materials. Routine materials testing functions, such as density, porosity, and elastic properties, can now be carried out easily and reliably by new nondestructive methods.

Ultrasonic NDC techniques can also be adapted to *in-situ* compaction, cold isostatic pressing, HIPing, and microwave process monitoring. We believe that the scope of our work will be further enhanced by its applications in QC/QA and other materials processing aspects of the industry.

Table 4. Ultrasonically determined properties of selected ceramics and composites.

MATERIAL	VELOCITIES (m/s)		MODULUS (GPa)			POISSON'S RATIO	TRANSDUCER F (MHz)	
	Long.	Shear	Young	Shear	Bulk		L.	Sh.
SiC (Dense)	11,820	7,500	397	180	200	0.170	20	10
SiO_2 (Slip Cast refractory)	4,140	2,740	30	13	13	0.110	1	1
Float Glass	5,815	3,460	72	30	44	0.230	10	10
BeO (Dense)	12,190	7,360	396	162	227	0.210	20	10
$Al_2O_3+ZrO_2+SiO_2$ (Refractory)	6,880	4,210	158	65	88	0.200	2	1
Graphite (Isotropic)	3,075	1,610	12.5	4.8	10	0.290	5	2
C-C (2D Comp. *Along fibers)*	7,500	—	100	—	—	—	2	1
C-C (2D Comp. *Across fibers)*	1,750	—	8.0	—	—	—	1	0.5
Diamond (Ind.)	18,770	12,100	1,176	514	551	0.144	50	20

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