MICROSTRUCTURE CHARACTERIZATION OF SUPERCONDUCTORS BY WIDEBAND ULTRASONIC SPECTROSCOPY

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Microstructure Characterization of Superconductors by Wideband Ultrasonic Spectroscopy

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ABSTRACT

Frequency dependence of ultrasound attenuation is a sensitive function of composition and microstructural characteristics of materials. For example, while a dense material of a given composition may transmit relatively broad spectrum of ultrasound, its coarse-grained, porous, or defective counterpart may only transmit certain selected frequencies. Investigation of frequency domain spectra can nondestructively provide significant process and applications related information about materials.

This paper provides an introduction to Wideband Ultrasonic Spectroscopy (WUS) by utilizing incident ultrasound - characterized by near "white" frequency spectra - and its applications for ceramic superconductors' NonDestructive Characterization (NDC).

Since ceramic superconductors are susceptible to damage by moisture and conventional wet coupling ultrasonic NDC techniques, special dry coupling transducers have been employed in this investigation. Preliminary results of dry coupling WUS are reported for YBC-based single phase and microstructurally heterogeneous superconducting compositions.

INTRODUCTION

Analogous to other spectroscopical methods that utilize optical, x-ray, γ-ray and other means of radiation for the measurement of their absorption through materials, ultrasonic frequency attenuation phenomenon can also be applied to the characterization of materials, Bhatia (1967).[1] Recent advances in structural and electronic materials have triggered unprecedented interest in nondestructive characterization.[2,3,4] Investigation of ultrasonic frequency domain spectra has been shown to reveal significant microstructural information about various materials, Serbian (1980)[5] and Generazio (1985).[6] More recently, ultrasonic methods have been extended for the NDC of ceramic superconductors in our laboratories,[7] and at NASA Lewis Center.[8]

While NDC methodolgy has been practiced for nearly 70 years, until recently it was restricted by the wet transducer coupling mechanisms. For obvious reasons, wet coupling techniques cannot be reliably used for the characterization of liquid-sensitive and microstructurally heterogeneous materials. Since the recent development of dry or self coupling transducers (up ~30MHz), it has been shown that liquid-sensitive electronic, green, and porous materials can be reliably and accurately characterized by modern ultrasonic methods, Bhardwaj (1987, 90).[9,10]

However, in order to conduct wideband ultrasonic spectroscopy, dry coupling methodology is not enough. A prerequisite of this method is exciting the test material with the "broadest" possible frequency spectrum so that at a given time the material's incident frequency response/selection can be evaluated over a relatively large composition of frequencies. This concept is analogous to *"white"* frequency spectra in optical spectroscopy. In ultrasound this condition is satisfied by critically damped and specific acoustic impedance matched transducers, Ultran (1982).[11]

In order to facilitate the NDC of ceramic superconductors we have devised special dry coupling delay line transducers (known as λ-series) with nominal frequencies >30MHz. Typical time and frequency domain characteristics for a 10MHz transducer used in this investigation are shown in Fig. 1.

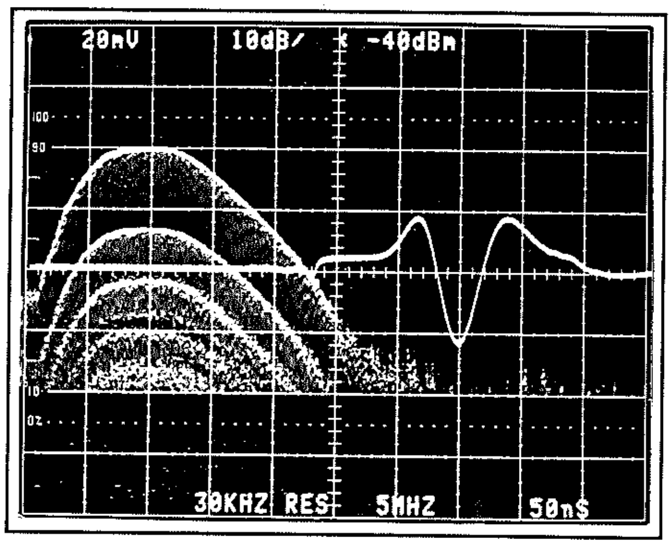
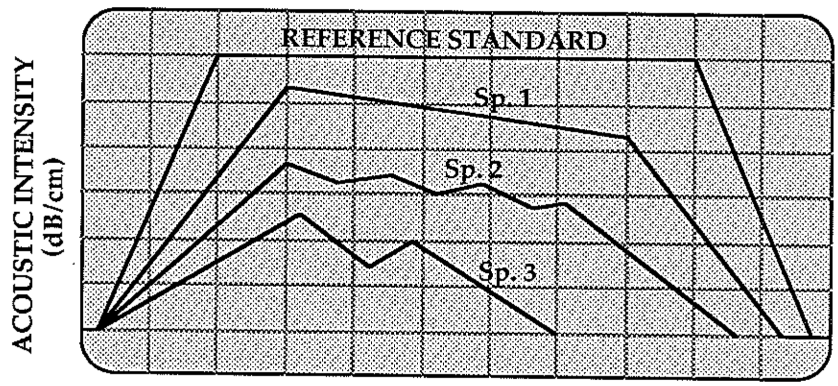


Fig. 1. Time and frequency domain characteristics of a dry coupling λ-series transducer - 6.0mm active area diameter and nominally 10MHz frequency - obtained from a 1.0cm optically flat and clear fused quartz.
Measured frequency domain parameters (from frequency spectrum).
Horizontal scale: 5MHz/div.; vertical scale: 10dB/div.
Bandwidth center frequency: 10.5MHz
Bandwidth at -6dB level: 3.5 to 17.5MHz or 140% of the center frequency
Bandwidth at -20dB level: 1.5 to 22.5MHz or 210% of the center frequency.
Measured time domain parameters (from horizontal trace).
Horizontal scale: 50ns/div.; vertical scale: 20mV/div.
Pulse width: ~90ns

The proposed WUS method utilizes a reference ultrasonic spectrum obtained by applying wideband transducers in direct transmission mode from a 1.0cm thick optically flat and polished clear fused quartz. Once the reference spectrum has been generated, similar spectra is obtained from the test materials by sandwiching them between the transmitting and receiving transducers. Assuming that the reference spectrum corresponds to input frequency components, subtraction of test materials spectra from the reference spectrum establishes relative frequency dependence of ultrasonic attenuation as a function of varying materials characteristics. An idealized process of this method is shown in Fig. 2.



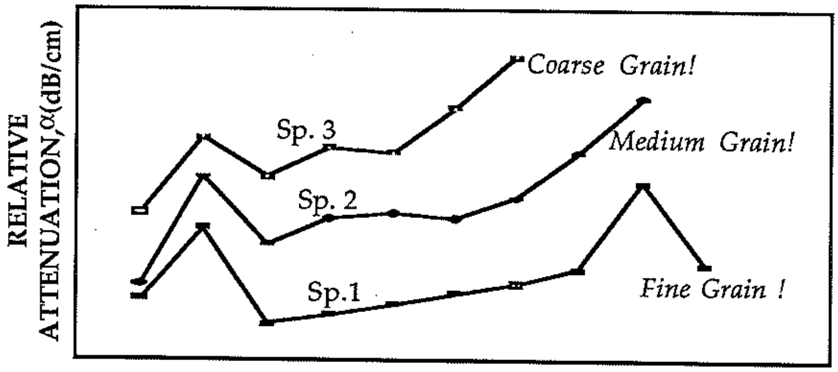


Fig. 2. An idealized sequence for Wideband Ultrasonic Spectroscopy (WUS).
TOP: Measured frequency domain spectra of the reference standard and test specimens, sp. 1, sp. 2, and sp. 3.
BOTTOM: Relative frequency attenuation obtained by subtracting the specimen spectra from that of the reference spectrum. Interpretation of specimens is provided for reference purpose only.

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EXPERIMENTAL PROCEDURE

In order to conduct WUS, the transmitting transducer - equivalent to characteristics shown in Fig. 1 - is excited by a short duration (~5ns) electrical pulse of controlled repetition frequency and the transmitted signals are received by the receiving transducer (with characteristics similar to that of the transmitting transducer). Transmitted signals are amplified by a broadband (~1KHz to ~50MHz) amplifier. The dry coupling transducers are clamped with test material sandwiched between them in order to maintain the constancy of pressure. The transmitted time domain signal corresponding to the propagation of ultrasound through the test material is converted into its frequency characteristics by analog or digital FFT (Fast Fourier Transformation) mechanism.

Fig. 3 shows the schematic representation of the ultrasonic system and transducers configuration with respect to test samples used in this investigation.

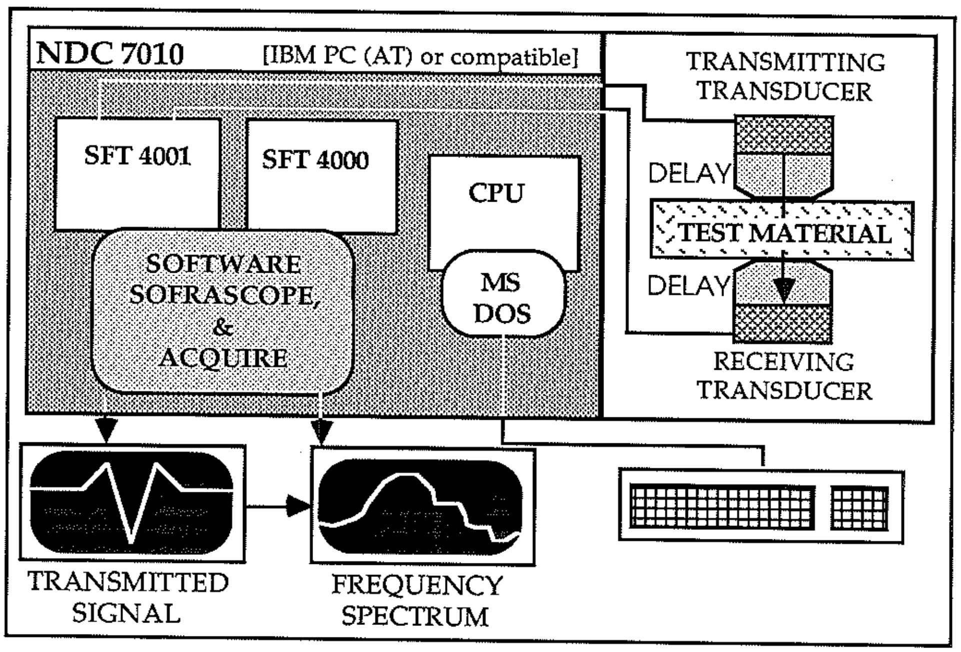

Fig. 3. Pulsed ultrasonic setup used for the measurement of frequency domain characteristics by sandwiching the test material between transmitting and receiving dry coupling highly broadband ultrasonic transducers.

SFT 4001: An ultrasonic pulser-receiver PCB
SFT 4000: An analog to digital converter PCB
SOFRASCOPE: Software for SFT 4001 set up
ACQUIRE: Software for time and frequency domain analysis.

It is important that once the reference spectrum from 1.0cm clear fused quartz has been obtained, then the pulser, amplifier, or FFT mechanism settings are not to be altered during the data acquisition from test materials.

WUS FEASIBILITY

In order to exhibit the feasibility of WUS, several samples of a diphasic material (1.0cm thick tungsten powder, varying in particle size, and compressed in a polymer matrix) were investigated by the procedure described here. Fig. 4 clearly shows that samples varying in particle size from 1 to 7µm exhibit relatively lesser frequency attenuation then the 30µm tungsten particle composite. Utilization of higher incident frequencies (~10 to 20MHz) generated enough resolution of smaller particle samples for further distinction of their microstructure.

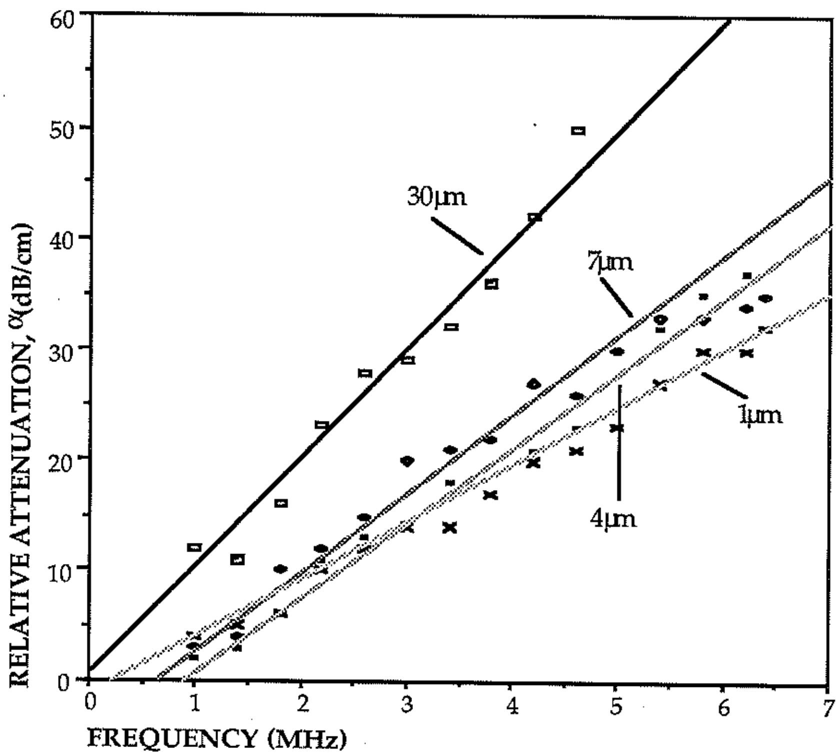

Fig. 4. Feasibility of Wideband Ultrasonic Spectroscopy established by frequency-dependence of ultrasonic attenuation by tungsten-polymer composite, varying in particle size.

OBSERVATIONS AND RESULTS FROM SUPERCONDUCTORS

Four YBC-based superconducting samples, 2 each corresponding to single phase and 1-2-3 glassy phase, were analyzed by the procedure described in the previous sections.

Table I shows the compositional and microstructural characteristics of these samples. Chemistry and methods of oxide superconductors are described by Bhalla, Roy, & Cross (1988).[12]

TABLE I. Characteristics of superconducting samples investigated by Wideband Ultrasonic Spectroscopy.

SN	COMPOSITION	GRAIN SIZE	DENSITY	PROCESSING PARAMETERS	SUPERCON-DUCTIVITY (Tc)
YBC-2	High purity phase pure orthorhombic	Fine	>90%	Sintered at 950°C, annealed at ~700°C in O_2, slow cooling	93°K
YBC-3[1]	Commercial single phase	Fine	~85%	Unknown	87°K
YBC-4[2]	70% YBC and 30% low melting glass (~450°C)	Medium to Coarse	---	---	Superconducting composite (levitation method)
YBC-5	Green phase (211) + YBC (123)	Coarse	High	Hot pressed at 900°C and 3500psi	Poor

[1]*Courtesy NASA Lewis Research Center, Cleveland, Ohio.*
[2]*Evidence of fractured interfaces between grains.*

Typical frequency spectra as a function of measured acoustic intensity of representative single and multi-phase superconducting samples are shown in Fig. 5 along with those of the reference clear fused quartz and copper. Since the test materials vary in thickness, their frequency spectra were normalized to yield db/cm units.

According to Fig. 5 relative to the reference spectrum, the single phase YBC superconductor exhibits lesser attenuation than its multi-phase counterpart, indicating higher scattering (due to larger grains) of ultrasound by the latter. Relative frequency attenuation for each sample was determined by subtracting its measured acoustic intensity from that of the reference clear fused quartz spectrum.

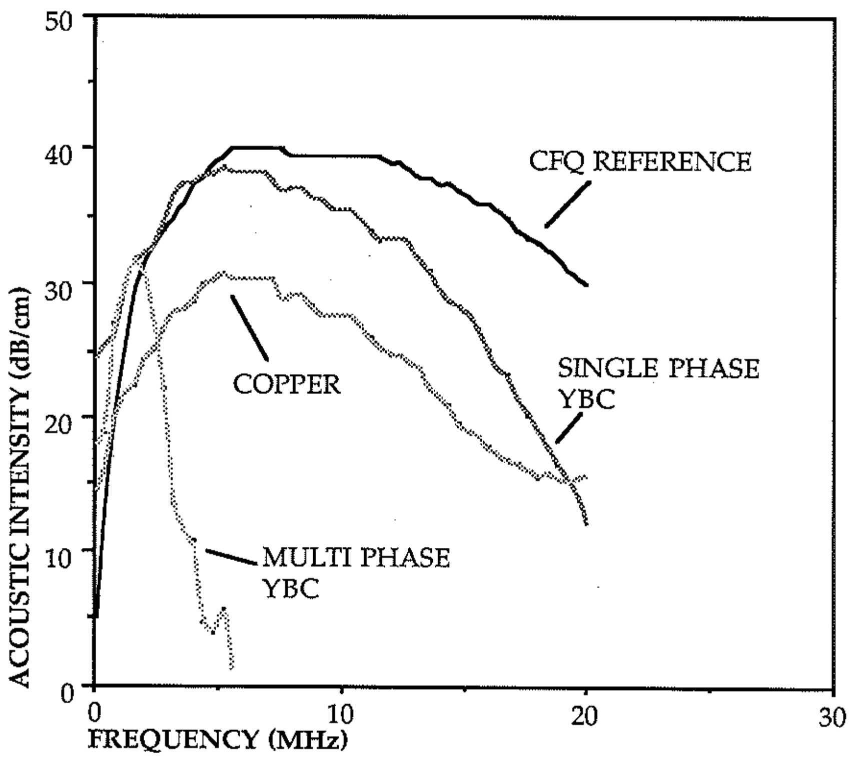

Fig. 5. Frequency domain spectra of single and multi-phase YBC-based superconductors shown along with the reference clear fused quartz spectrum and a commercial sample of copper.

Spectroscopy of single phase compositions

Fig. 6 shows the relative frequency attenuation for single phase YBC-2 and YBC-3 samples along with that of copper as a reference.. The trend of attenuation for both samples is similar, however, numerically YBC-2 exhibits lesser frequency attenuation than YBC-3. Since frequency attenuation in polycrystalline materials of the same chemical composition increases with grain size and decreases with density, it can be concluded that either YBC-2 is a finer grain material or it is denser than the YBC-3 sample.

Assuming the constancy of composition, longitudinal wave velocity measurements can also provide significant information about test material densities.[9] Higher velocities are indicative of higher densities. In the present case the longitudinal wave velocity of YBC-2 is 4600m/s, while for YBC-3 it is 4500m/s, supporting the argument that the former is denser than the latter.

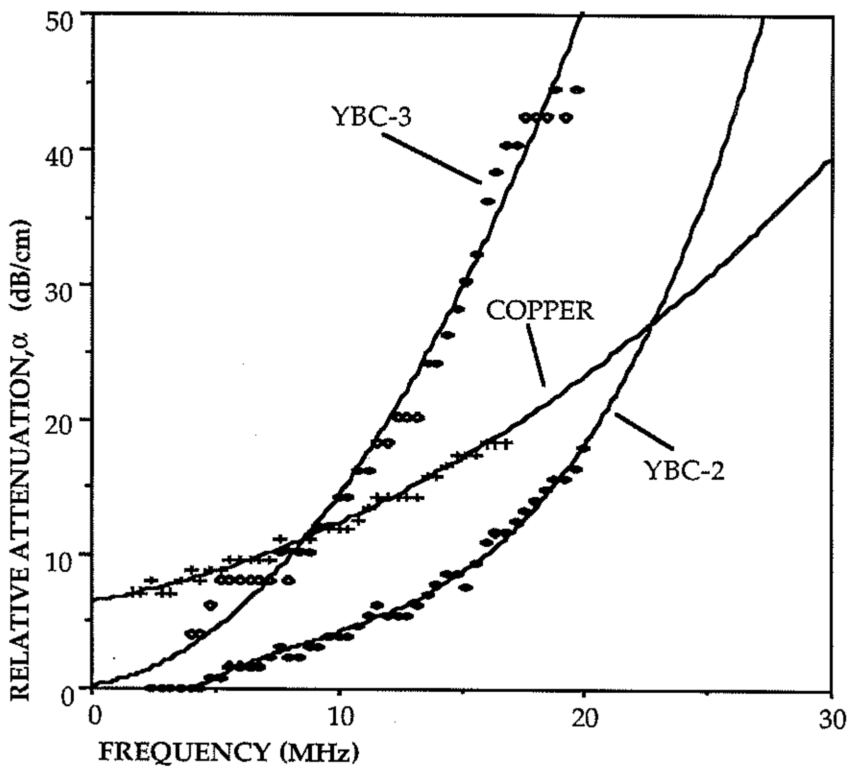

Fig. 6. Relative frequency attenuation of single phase superconducting samples shown along with that of copper for reference purpose.

Spectroscopy of multi-phase compositions

Figure 7 shows relative frequency attenuation of multi-phase superconducting samples, indicating higher frequency attenuation of YBC-5 than YBC-4. Assuming that texture plays the dominant role in frequency attenuation of these samples, it can be concluded that YBC-4 is a finer grained material than the YBC-5.

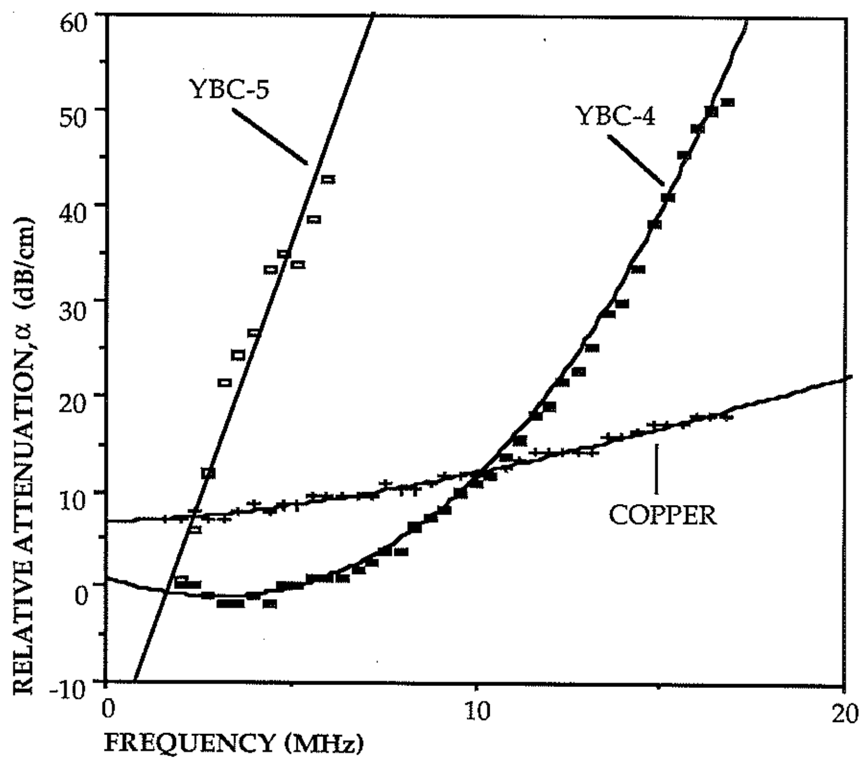

Fig. 7. Relative frequency attenuation of multi-phase YBC superconducting samples shown along with that of copper for reference purpose.

CONCLUSIONS

In this paper we have introduced Wideband Ultrasonic Spectroscopy (WUS) as a significant tool for the NDC of materials. We have further established the feasibility of the dry coupling ultrasonic mechanism for reliable characterization of liquid-sensitive material such as ceramic superconductors.

Our preliminary observations from WUS of single and multi-phase superconducting samples indicate a definite relationship that apparently describes frequency-dependence of ultrasonic attenuation as a function of test material microstructure. However, in order to gain more knowledge about the bulk characteristics and composition by WUS, it is imperative that selectively varying compositions and processing conditions of superconducting materials be examined by this method. Besides the obvious advantages of ultrasonic NDC, it is believed that systematically determined time and frequency domain data from superconductors might be directly related to an applications oriented property, such as the definition of T_C.

ACKNOWLEDGMENTS

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