Experimental Study of Relationships between Ultrasonic Attenuation and Dispersion for Ceramic Matrix Composite

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Abstract

In this paper an experimental study of different ceramic matrix composites with high elastic losses and dispersion (porous piezoceramics, composites ceramics/crystals) were carried out. Complex sets of elastic, dielectric, and piezoelectric parameters of the porous piezoceramics and ceramic matrix piezocomposites were determined by the impedance spectroscopy method using Piezoelectric Resonance Analysis software. Microstructure of polished and chipped surfaces of composite samples was observed with the optical and scanning electron microcopies. Experimental frequency dependencies of attenuation coefficients and ultrasonic velocities for different ceramic matrix composites were compared with the theoretical results obtained using general Kramers-Kronig relations between the ultrasonic attenuation and dispersion.

Keywords: elastic losses; scattering; dispersion; ceramic matrix piezocomposites; complex impedance.

1. Introduction

The multiphase ceramic matrix composites are very complex objects for theoretical modelling, NDT (non-destructive testing), and ultrasonic measurements (Rybyanets et al., 2011). The accurate description of piezocomposites must include the evaluation of the dielectric, piezoelectric and mechanical losses, accounting for the out-of-phase material response to the input signal (Rybyanets et al., 2007a). It was shown (Nasedkin et al., 2005) that pulse-echo measurements of frequency dependencies of elastic properties for dispersive and lossy ceramic composites are inaccurate and ambiguous. In its turn, piezoelectric resonance measurements (PRAP) can give accurate and reproducible results that well agree with the results of 3D finite-difference simulations.

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Different methods were proposed for theoretical modelling and evaluation of ceramic composites properties (Nasedkin et al., 2005). However, physical mechanisms and interrelations of elastic losses and dispersion in complex objects, such as, porous ceramics and ceramic matrix piezocomposites remain difficult to deeply understand and they would require a dedicated study.

In this paper the frequency dependencies of complex elastic moduli for different ceramic matrix composites with strong spatial dispersion and high elastic losses were measured and compared with the theoretical predictions.

2. General Relationships between Ultrasonic Attenuation and Dispersion

Relationships between attenuation and dispersion, sometimes called Kramers-Kronig or generalized dispersion relationships, have proved useful in several areas of physics (Mangulis, 1964).

Expressed in a form appropriate to ultrasonic studies, these relationships take the form (the derivations of the general relations are readily available elsewhere (O’Donnell et al., 1981):

$$ K_1(\omega) - K_1(\infty) = \frac{2}{\pi} \int_0^\infty \frac{K_2(\omega') d\omega'}{\omega'^2 - \omega^2}, $$

$$ K_2(\omega) = -\frac{2\omega}{\pi} \int_0^\infty \frac{K_1(\omega') - K_1(\infty)}{\omega'^2 - \omega^2} d\omega', $$

where $K_1(\omega)$ and $K_2(\omega)$ are the real and imaginary parts, respectively, of the dynamic compressibility (inverse of the bulk modulus). If the ultrasonic wave vector is written as $k = \frac{\omega}{c(\omega)} + i\alpha(\omega)$, then $C(\omega)$ is the phase velocity and $\alpha(\omega)$ is the attenuation coefficient for the incident wave, as observed in transmission (i.e., direct "straight-line" propagation) measurements.

In the form given by Eqs. (1) and (2) the Kramers-Kronig relationships are limited in usefulness because of their nonlocal character; i.e., a knowledge of either the attenuation or the dispersion for all frequencies is required. More useful, approximate nearly local forms of the attenuation-dispersion relationships can be obtained from the exact nonlocal forms given in Eqs. (1) and (2) under the assumptions that the attenuation and dispersion are sufficiently small and do not change rapidly over the frequency range of interest. These nearly local relationships are:

$$ \alpha(\omega) \approx (\pi \omega^2 / 2 C_0^2) \frac{dC(\omega)}{d\omega}, $$

$$ \Delta C(\omega) = C(\omega) - C_0 \approx \frac{2 C_0^2}{\pi} \int_0^\omega \frac{\alpha(\omega)}{\omega^2} d\omega', $$

where $\omega_0$ is some convenient reference frequency and $C_0 = C(\omega_0)$ is the phase velocity at this reference frequency. The validity of these approximate relationships was demonstrated in several acoustic systems exhibiting substantially different attenuation and dispersion mechanisms (Mangulis, 1964; O’Donnell et al., 1981).

3. Experimental Procedures

3.1. Experimental samples

The following two types of ceramic composites (Rybyanets et al., 2011; Rybyanets et al., 2007b) with high losses were chosen as model samples for simulation of ultrasonic wave propagation and comparison with piezoelectric resonance analysis (PRAP) and ultrasonic measurements (Figure 1):

- ceramic matrix composites A850L (Figure 1a) consisting of soft PZT matrix with randomly distributed α-Al2O3 crystals with a mean particle diameter ~ 200 μm and volume fraction from 9 up to 26 vol.%;
- porous PZT piezoceramics PCR-1 (porosity 18 %, average pore size ~ 20 μm) shown in Figure 1b.
3.2. Measurement methods

Complex elastic, dielectric and piezoelectric coefficients of porous ceramics and ceramic matrix piezocomposite elements were determined by impedance spectroscopy method using PRAP software (Rybyanets et al., 2007b; Rybyanets et al., 2013). Measurements were made using Agilent 4294A Impedance Analyser. The microstructure of polished and chipped surfaces of composite samples was observed with optical (NeoPhot-21) and scanning electron microscopes (SEM, Karl Zeiss).

4. Results and Discussion

Figure 2a shows the frequency dependencies of ultrasonic velocities \( V_t^D = \sqrt{C^D_{33}/\rho} \) and \( V_t^E = \sqrt{C^E_{33}/\rho} \) and corresponding attenuation coefficients \( \alpha(V_t^D) = C^D_{33} \omega_0 / 2C^D_{33} \) and \( \alpha(V_t^E) = C^E_{33} \omega_0 / 2C^E_{33} \) obtained from PRAP results the higher harmonics of the TE mode of PCR-1 porous ceramic disks with different fundamental resonant frequencies. The following standard notations were used above: \( C^D_{33}, C^E_{33} \) - real and \( C^D_{33}^I, C^E_{33}^I \) – imaginary parts of elastic stiffness, \( \rho \) – density, \( \omega_0 \) - cyclic frequency.

The least squares approximations of the frequency dependencies of Figure 2a have shown that in the frequency range corresponding to Rayleigh scattering of high frequency ultrasonic waves on pores (\( \lambda >> D \), where \( D \) – average
pores diameter, \( \lambda - \) wavelength), attenuation coefficients \( \alpha(V_t^D) \) and \( \alpha(V_t^E) \) grow with the frequency approximately as \( f^4 \). At further frequency growth scattering mechanism changes from Rayleigh type to stochastic one (\( 4 \leq \lambda/D \leq 10 \)) and frequency dependencies of \( \alpha(V_t^D) \) and \( \alpha(V_t^E) \) are approximated by \( f^4 \) function. The corresponding frequency dependence of ultrasonic velocity \( V_t^D \) defined by the elastic modulus \( C_{33}^D \) is approximated by \( f^4 \) function (normal dispersion type) in Rayleigh range and by linear function that agreed with the theoretical predictions of Section 2 for the dispersion (Equations 3 and 4). In its turn, ultrasonic velocity \( V_t^D \) defined by the elastic modulus \( C_{33}^D \), decreases with the frequency (anomalous dispersion) as result of the electromechanical contribution to \( C_{33}^E \) according the equation \( C_{33}^D = C_{33}^E(1-k_t^2) \) (note that effective value of \( k_t \) measured on higher harmonics drops drastically with the harmonics number as \( k_t^2 = \frac{8k_t^2}{(2n+1)\pi^2} \)).

Figure 2b shows the frequency dependencies of ultrasonic velocities \( V_t^D = \sqrt{C_{33}^D/\rho} \) and corresponding attenuation coefficients \( \alpha(V_t^D) = C_{33}^D \omega_0/2C_{33}^D \) for ceramic composites PZT/\( \alpha-\text{Al}_2\text{O}_3 \) with different volume fractions of \( \alpha-\text{Al}_2\text{O}_3 \) measured on standard disks samples by piezoelectric resonance (PRAP) method. The results of complex material constants evaluation are well agreed with the previously reported results for lossy piezoelectrics (Rybianets et al., 2011; Rybianets, 2011). The least squares approximations of the frequency dependencies of Figure 2b have shown that in all measured frequency range and for all concentration of \( \alpha-\text{Al}_2\text{O}_3 \) particles, attenuation coefficients \( \alpha(V_t^D) \) and \( \alpha(V_t^E) \) grow with the frequency as \( f^4 \) and corresponding frequency dependence of ultrasonic velocity \( V_t^D \) is approximated by linear function. This behavior corresponds to stochastic type of scattering (\( 4 \leq \lambda/D \leq 10 \)) and is described well by the approximate dispersion relations (Equations (3) and (4)). It can be mentioned that unlike from \( V_t^D \) behavior measured on higher harmonics for porous ceramics PCR-1 the dispersion of ultrasonic velocity \( V_t^D \) measured on fundamental resonant frequency has normal character, because of constancy of electromechanical contribution (\( k_t \) to \( C_{33}^D \)).

5. Conclusion

An experimental study of different ceramic matrix composites with high elastic losses was carried out. The strong spatial dispersion was found. Experimental frequency dependencies of the attenuation coefficients and ultrasonic velocities for porous ceramics and ceramic/crystal composites were compared with the theoretical results obtained using general Kramers-Kronig relations. It was shown that depending on the frequency range and scattering particles size the scattering mechanism can changes from Rayleigh (\( \lambda >> D \)) to stochastic type (\( \lambda \sim D \)) that leads to corresponding changes in the dispersion character and well corresponds to the theoretical predictions.

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