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Measuring the elastic tensor of a monolithic SiC hollow cylinder with Resonant Ultrasound Spectroscopy

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Abstract

Resonant Ultrasound Spectroscopy (RUS) has become a standard tool in material science to measure nondestructively the elastic properties of materials. The widespread adoption of this tool for applications related to nondestructive testing and structural health monitoring in the industry and other places has been restricted thus far by some requirements imposed by RUS on the sample to be tested, in particular the fact that the sample geometry should be relatively simple (e.g., cuboid, cylinder, or sphere). In this paper, we use RUS to determine the baseline material properties of a hollow cylinder made of monolithic SiC, a surrogate material and geometry representing nuclear fuel cladding for this study. Typically, elastic properties of the nuclear fuel cladding samples are inspected destructively (e.g., indentation [1]) or with acoustic techniques

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[2] requiring multiple and cumbersome measurements. Here, a relatively new RUS approach specifically developed to measure the elastic tensor of samples with arbitrary geometries is used. The results obtained from this RUS approach are compared to values reported in the literature. As a second verification step, vibrational mode shapes computed numerically with the elastic constants are compared to those measured on the sample using a 3D laser Doppler vibrometer, showing an excellent agreement.

1. Introduction

Resonant Ultrasound Spectroscopy (RUS) has emerged as a fast, inexpensive, and accurate method for measuring the elastic properties of a material [3-5]. The first implementation of RUS can be traced back to seismological applications and the measurement of the Earth elastic properties, where the phase velocity within were estimated from its free oscillations after a large earthquake [6]. Since then, the technique has been refined and used in a wide range of applications including material science, geophysics, biomedical engineering, civil engineering, and MEMS [7-9].

RUS starts with a set of experimentally measured resonance frequencies for a given specimen. The set of frequencies is uniquely determined by the geometry of the specimen, the elastic tensor of the material, its crystallographic orientation, its mass density, and its damping. Determining the elastic tensor of the material using the measured resonance frequencies is a two step process in which the resonance frequencies predicted by a numerical or analytical model (forward step) are compared to those measured experimentally and the elastic constants of the material are adjusted accordingly in the model to minimize the squared difference between measured and calculated frequencies (inversion step). Typically, the forward step involves the application of the Rayleigh-Ritz method [10], which provides the eigensolution in the form of truncated series of analytical functions. These functions can in turn be used by a minimization algorithm that requires a continuous search space during the inversion step, which is readily available from the summation of analytical functions used in the forward problem. A brief literature review indicates that the Levenberg-Marquardt method [11], which is essentially a hybrid of gradient-descent and Gauss-Newton methods, has been the optimization algorithm of

choice for the RUS inversion step. This particular combination of a semi-analytical solution obtained from the forward problem and inversion algorithm is limited to samples with relatively simple geometries (i.e., cuboids, cylinders, and spheres). Recently, RUS has been extended by Remillieux et al. [12] to samples with arbitrary geometries. They achieved this by solving the forward problem with the finite-element method (FEM) and the inversion problem with an algorithm capable of handling discrete solution spaces. They selected and implemented a genetic algorithm for the inversion step and validated the approach against numerical and experimental data. The advantage of their approach over the traditional RUS approach is the ability to find a global minimum within a wide search space, without a need for an initial guess that would be relatively close to the sought solution.

The motivation behind this work is to determine if RUS could be used as a practical tool to monitor the evolution of damage in nuclear fuel-cladding. Previously, the elastic moduli of SiC have been measured with pulse-echo ultrasonics, tensile tests, flexural tests, nano-indentation, Vicker's indentation, and ring compression, with an extensive review of these techniques provided by Snead et al. [13]. In a tensile or flexural test, the specimen is prone to failure due to the brittle nature of SiC and the pressure applied by the grips at the boundaries of the specimen. Besides, these tests can only measure one elastic constant (as opposed to the full elastic tensor). The ring compression technique consists of internally pressurizing or diametrically loading the sample until failure is achieved [14]. Indentation techniques can measure fracture toughness, elastic modulus, and hardness but they do so locally (as opposed to a global measure for the entire specimen) and at the expense of ablating the surface of the specimen. Given that the end goal of our research effort is to monitor the evolution of damage in a fuel-cladding specimen, the technique used to measure the elastic constants should be non-

destructive. Pulse-echo ultrasonics is a non-destructive technique measuring the propagation speed of elastic waves (compressional and shear waves) in the specimen. For an isotropic material, the compressional and shear waves are correlated to the elastic constants C_{11} and C_{44}, respectively. However, applying this technique on specimens with complex geometries and/or anisotropy is not trivial. A specimen made of SiC in intact condition is expected to be isotropic. However, as damaged is progressively induced by the environment (e.g., radiation and temperature), the material may become anisotropic and experience softening (i.e., a drop of the elastic constants) [15, 16]. In principle, RUS can measure the full elastic tensor of a material with one measurement and so it can monitor material softening and evolution of anisotropy simulatenously without further damaging the sample. The RUS approach proposed by Remillieux et al. [12] was shown to be applicable to arbitrary geometries. Is it applicable to fuel cladding?

In this paper, the elastic properties of a thin and hollow cylinder in pristine condition made of monolithic SiC, which is a surrogate for nuclear fuel cladding, are measured using the RUS approach proposed by Remillieux et al. [12]. This is a first step towards monitoring the evolution of the elastic properties (and consequently material integrity) as a fuel-cladding specimen is exposed to harsh conditions. To validate the new RUS approach, we compare (a) the elastic properties to values reported in the literature using other methods and (b) the computed mode shapes to the ones measured experimentally using a non-contact laser Doppler vibrometer.

2. Experimental Setup

RUS experiments were conducted on a monolithic SiC sample to estimate its elastic properties. The sample consists of a hollow cylinder with a length of 16.03 mm, outer radius of 4.26 mm, inner radius of 3.56 mm, and a mass density (assumed uniform) of 3208 $kg.m^{-3}$. The weight and dimensional measurements were conducted in our laboratory, with an error for dimensional measurements smaller than 0.005 mm. It was manufactured by Dow Chemical Company out of vapor-deposited (CVD) silicon carbide (SiC). The experimental setup and sample are shown in Fig. 1a and b, respectively.

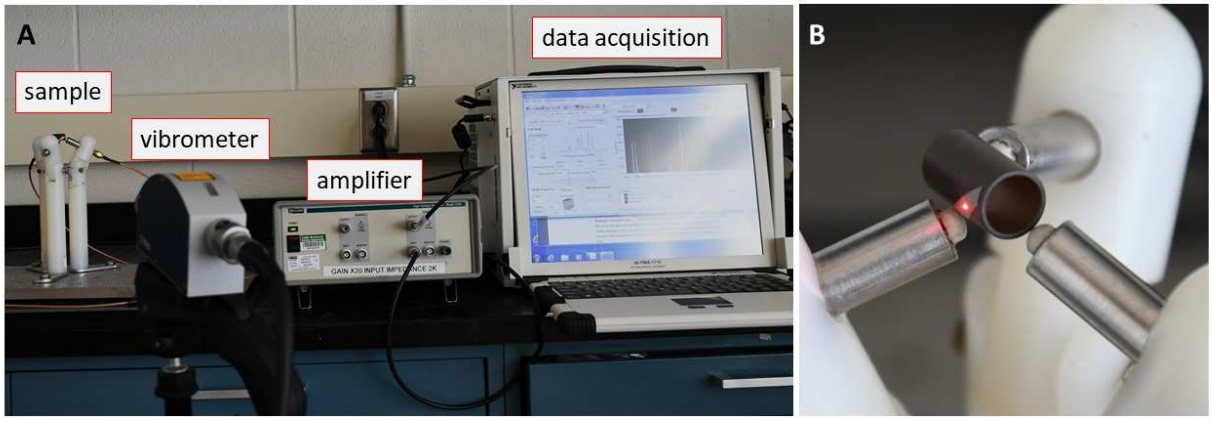

Figure 1: Photographs of (a) experimental setup and (b) close-up of the SiC sample with the laser beam focused on reflecting tape.

The setup used to probe the sample includes the stand made of three small supports shown in Figure 1b. The supports have a small surface area in contact with the cylinder such that free (unconstrained) boundary conditions can be approached while providing the necessary source of vibrational excitation. The supports are piezoelectric transducers capped with ceramic, with only one acting as a source and the other two being turned off to provide support to and

stabilize the sample. The vibrational response of the sample was measured at one point with a laser Doppler vibrometer (Polytec OFV-301) focused on a small strip of reflecting tape. The source transducer was driven by a function generator (National Instrument PXI-5406) transmitting a 4V peak-to-peak signal to a 20× gain voltage amplifier (TEGAM 2350). The output of the laser vibrometer was digitized by a data acquisition card (National Instrument PXIe-5122) with sampling rate of 10MHz. The RUS experiment was repeated 9 times with the sample being repositioned on the stand between the tests. In all experiments, the source transducer was digitally swept up with a sequence of harmonic voltage signals, ranging between 50 and 400 kHz in frequency steps of 10 Hz. At each step, a harmonic signal was generated for 50 ms and the vibrational response was recorded during the last 30 ms of excitation to ensure that steady state conditions had been reached. Vibrational spectra were constructed from the harmonic responses, using heterodyne processing. Once the spectra have been measured, resonance frequencies and quality factors (a measure of the material damping) are extracted by fitting a Lorentzian function around each resonance peak. The vibrational spectrum can be described by a sum of Lorentizian line shapes with amplitudes a_k,

$$Y(f) = \sum_{k=1}^{M} \frac{a_k}{\left(f_k^2 - f^2\right) + i\left(f_k f / Q_k\right)} \tag{1}$$

Where f_k and Q_k are the undamped resonance frequency and quality factor for the k^{th} mode, respectively.

3. RUS inversions

The nine measured amplitude spectra of the SiC sample are shown in Figure 2. The figure shows the magnitude of vibration of the sample over a wide frequency range. The location of the resonance frequencies are denoted by vertical black lines and are additionally given in Table 1. Given the symmetry of the sample, degenerate modes are expected when the sample vibrates with a bending type of motion. Due to imperfections in the sample geometry, boundary conditions (the sample is not exactly unconstrained when resting on the stand), and possibly the mass distribution (the mass is not necessarily uniformly distributed), most degenerate modes lead to a set of two distinct resonant frequencies that can be individually extracted. Note that since the RUS algorithm assumes perfect symmetry and unconstrained boundary conditions, the forward step of the algorithm will lead to a set of two identical frequencies for each degenerate mode. The algorithm will eventually lead to a set of two identical frequencies lying in between the set of distinct experimental frequencies.

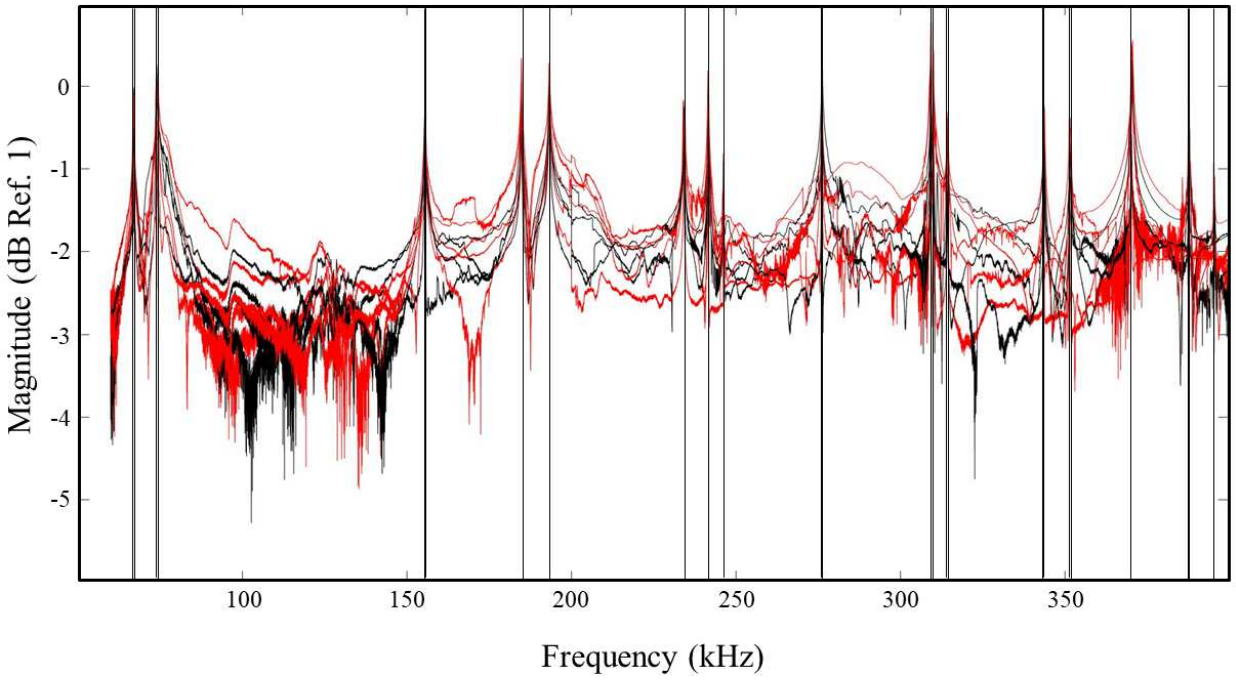

Figure 2: Resonant ultrasound spectra of the SiC sample, repositioned after each measurement. The back and red color for the curves are used for legibility and do not have a particular meaning.

The extracted set of frequencies served as input to the RUS algorithm developed at Los Alamos National Laboratory. The forward step of the RUS algorithm consists of computing the eigensolution (resonance frequencies and mode shapes) of the sample with free boundary conditions based on a given set of elastic constants. This step is achieved with the commercial finite-element software package COMSOL Multiphysics. In COMSOL, the computational domain was discretized into 40,148 quadratic tetrahedral elements. The maximum element size was set to 0.4 mm, which ensured that there were at least 10 elements per smallest wavelength at 400 kHz (for reference, the expected wavelength of a shear wave in this material at 400 kHz is λ_s

= 20 mm). Each eigenvalue problem (forward step) took a few seconds to solve. The objective of the inversion step is to find the set of elastic constants that will lead to the smallest difference between computed and measured resonance frequencies. The inversion step is achieved with a genetic algorithm developed in house, using the same selection rate (50%) and mutation rate (40%) as in the work of Remillieux et al. [12]. The finite-element model and the genetic algorithm were coupled and fully automated to achieve the RUS inversion. The eigenvalue calculations and inversions were carried out by a workstation equipped with two Intel Xeon CPU E5-2687W v4 12-core processors (clock speed of 3 GHz) and 512 Gb of RAM. The calculation time is highly dependent on the available computer resources but also on the convergence rate of the solution. In our case, the elastic constants could be computed within a few hours at most.

Table 1 lists the measured frequencies from RUS, frequencies calculated with FEM, and the *Q*-factors extracted from the spectra. Table 2 lists the evaluated elastic constants. Good agreement is observed between the computed and measured resonance frequencies, with a total RMS error for all the computed modes of 0.793%. Most resonance modes exhibit an error smaller than 1%. Errors larger than 1% are observed for modes 7 and 8, possibly as a result of their close proximity (i.e., modal overlap). Other possible sources of error include: i) the elastic tensor of the material could have a cubic symmetry instead of the isotropic symmetry assumed in this study, ii) non-uniform mass density, iii) geometry imperfections, iv) boundary conditions that are not perfectly unconstrained. The values for the elastic properties quantified in this work are consistent with previous measurements of SiC material. The value of the elastic modulus is clearly within the range set by methods such as four-point flexural bending and other sonic

methods [17, 18]. The value for the Poisson ratio is slightly smaller than the other reported values.

Table 1: Resonance frequencies (f_k) and Q-factors extracted from the spectrum and the associated computed resonance frequencies based on the combined genetic algorithm and FEM simulations. The symbol * indicates degenerate modes of the sample, and the superscript † indicates when those modes were independently resolvable.

Mode k	Extracted f_k (Hz)	Computed f_k (Hz)	Extracted Q
1	66,844.8	66,600.0 (-0.37%)	829.0
1*†	67,035.0	66,601.6 (-0.65%)	
2	74,029.8	74,644.4 (+0.83%)	363.7
2*	74,029.8	74,645.4 (+0.83%)	
3	155,428.2	154,279.9 (-0.74%)	426.8
3*	155,428.2	154,280.4 (-0.74%)	
4	184,796.6	184,384.5 (-0.22%)	1,607.0
4*	184,796.6	184,384.8 (-0.22%)	
5	193,218.0	194,880.0 (+0.86%)	1863.3
5*	193,218.0	194,880.5 (+0.86%)	
6	234,077.2	234,921.3 (+0.36%)	2,378.1
6*	234,077.2	234,922.2 (+0.36%)	
7	241,492.3	237,924.3 (-1.48%)	2,078.9
7*†	241,665.6	237,924.3 (-1.55 %)	
8	246,139.7	250,326.6 (+1.70%)	612.1
9	276,009.0	273,711.4 (-0.83%)	3738.7
9*†	276,188.1	273,712.3 (-0.90%)	
10	309,144.9	311,918.7 (+0.90%)	5828.7
10*†	310,066.1	311,918.7 (+0.60%)	
11	314,064.3	313,904.4 (-0.05%)	3525.3
11*†	314,197.9	313,905.7 (-0.09%)	
12	343,449.3	343,564.4 (+0.03%)	4397.6
12*†	343,604.6	343,588.6 (-0.00%)	
13	351,331.1	354,413.1 (+0.88%)	3043.4
13*†	351,598.4	354,437.5 (+0.81%)	
14	370,195.1	366,883.1 (-0.89%)	4751.8
15	387,510.1	389,113.3 (+0.41%)	4564.8
15*†	387,688.5	389,133.8 (+0.37%)	
16	395,148.1	392,029.8 (-0.79%)	2849.4
16*†	395,362.4	392,032.7 (-0.84%)	

Table 2: Free elastic moduli, C_{11} and C_{44}; Young's modulus, E; Poisson Ratio, v; bulk modulus, K; and the total RMS error for all modes.

	C_{11} [GPa]	C_{44} [GPa]	E [GPa]	v	K [GPa]	RMS Error
Computed	455.7 (±0.81%)	206.8 (±1.4%)	448.5	0.085	180	0.793 %
Yavuz [17]	-	-	431 (±5.6%) 4 pt flex	0.13 (±0.15%)	-	-
Dow Chemical Company [18]	-	-	466 sonic 461 4 pt flex	0.21	-	-
Singh [19]	-	-	439.8 (±1.0%) RUS	0.114 (±10.5%)	-	-

The RUS analysis was validated through a second set of experiment. Typically, the RUS analysis leads to a set of measured and predicted (with error analysis) resonance frequencies, if needed a set of measured quality factors, and a set of elastic constants (also with error analysis). If there are uncertainties about the symmetry of the elastic tensor and/or a high modal density, as this is the case in this particular analysis, the order in which the modes come for a given set of elastic constants might be incorrect. One way to verify this is to conduct a modal analysis and compare the experimentally measured mode shapes to the ones predicted using the elastic tensor from the RUS inversion. The mode shapes of the sample were measured without contact using a 3D laser Doppler vibrometer (Polytec PSV-3D-500) while the sample was resting on and excited by the 3-point-support stand depicted in Fig. 1b. We used the same excitation amplitude and frequency range as in the RUS experiment. A section of the sample surface spanning its length was scanned using 171 points. This section was covered with a very thin layer of retroreflective tape (which

can reflect the laser beam back to where it is emitted from up to grazing incidence) so that a large angle of the curved surface could be scanned by the laser system. The source signal was a chirp spanning the frequency range of interest. The measured and predicted mode shapes are shown in Fig. 3 for the first 16 modes. This figure unambiguously demonstrates that all mode shapes have been predicted correctly and in the correct order. Typically, for a RUS analysis, at least 5 times as many mode as there are elastic constants to invert for should be measured, so in this case 10. In this paper, we used 16 to increase the confidence of our analysis and obtain an excellent match for these 16 modes.

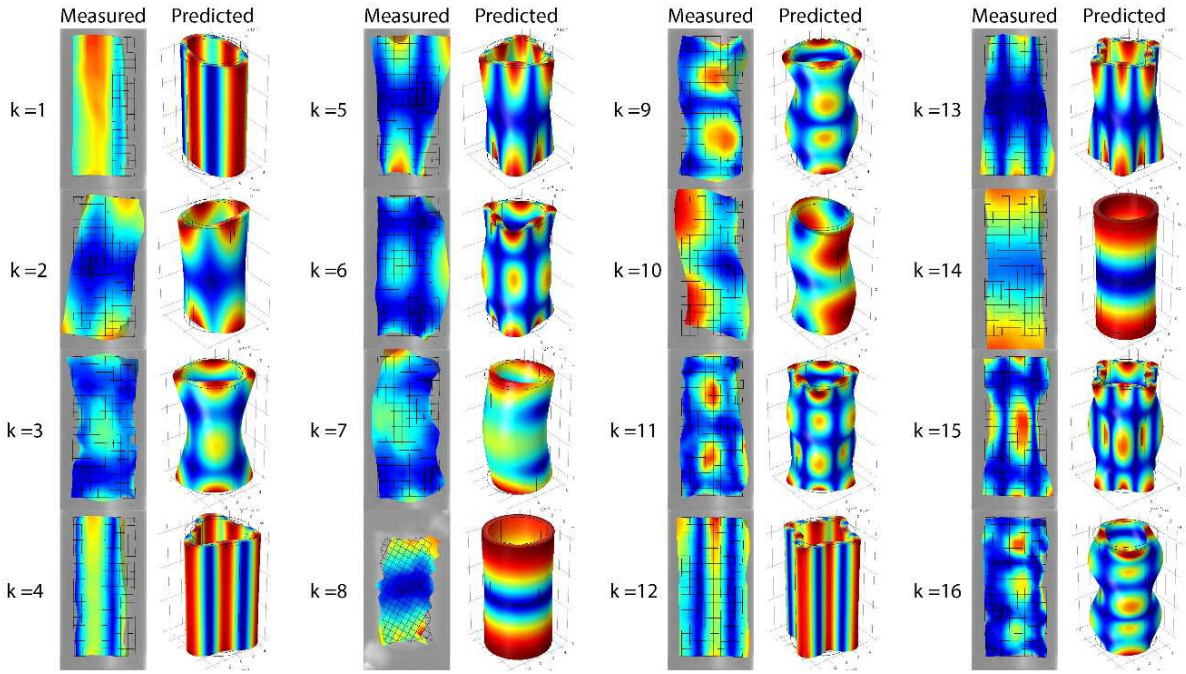

Figure 3: Comparison between the mode shapes of vibration measured with the 3D laser Doppler vibrometer and computed with FEM based on the elastic constants retrieved from the RUS inversion.

4. Conclusions

In this paper, the elastic properties of a thin hollow cylinder made of monolithic SiC were measured using a relatively new RUS approach developed by Remillieux et al. (2015). This approach is based on the combination of FEM simulations and a genetic algorithm for the inversion of the elastic constants. It was designed specifically to address the analysis of samples with arbitrary geometries. In the past, such measurements have been restricted by the non-trivial geometry of the sample. The elastic properties derived from this new approach were shown to agree well with previous measurements of SiC. Furthermore, the mode shapes of vibration for the first 16 modes simulated with FEM and the set of elastic constants computed from the proposed approach agree well with the experimental data collected with a 3D laser vibrometer. The successful demonstration that the elastic properties of a tubular SiC specimen can be determined accurately with RUS is a promising step towards the nondestructive inspection of fuel assembly core components, such as fuel cladding, that are exposed to harsh radiation and temperature conditions.

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