

Effects of Graphite Porosity and Anisotropy on Measurements of Elastic Modulus using Laser Ultrasonics

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Abstract—Laser ultrasonic techniques can be used to study the ultrasonic properties of nuclear graphites and can serve as tools in establishing relationships between materials microstructure and the macroscopic stiffnesses of graphite. Establishing structure-property relationships permits improved ultrasonic sensing of graphite microstructural changes related to service-induced degradation. Laser ultrasonic measurements were made using a pulsed Nd:YAG laser source and detection was performed using a Michelson-type interferometer. This source-receiver combination provides for non-contacting, highly linear transduction of broadband, ultrasonic pulses permitting simultaneous determination of longitudinal and shear stiffnesses. Measurements show that among the graphites examined, a change in density of 0.21 g/cm³ (average 1.8 g/cm³) results in a change in the longitudinal elastic stiffness of 7.1 GPa (average 12.2 GPa) and 3.2 GPa (average 4.3 GPa) for the shear stiffness. Larger variations in density were produced by controlled oxidation of IG-110 and NBG-18. Shear wave birefringence measurements using laser line sources in IG-110 and PCEA indicate that IG-110 behaves isotropically while PCEA displays texture characteristic of transversely isotropic materials.

Keywords—laser ultrasonics; graphite; porosity; Young's modulus; anisotropy; shear birefringence

I. INTRODUCTION

According to the *Nuclear Energy Research and Development Roadmap, Report to Congress*, data and methods are needed to assess the integrity of structures and components for safe and sustained nuclear plant operation [1]. This report details how long-term materials degradation must be understood and used to predict material behavior. In particular, a milestone for materials research and development is the baseline characterization of nuclear graphite. The ability to nondestructively assess the microstructural state of nuclear-grade graphite is critical to qualifying these materials for current and future nuclear reactors. Laser ultrasonic techniques can be used to study the ultrasonic properties of nuclear graphites and can serve as tools in establishing relationships between materials microstructure and the macroscopic stiffnesses of graphite. Variations in graphite porosity can be related directly to overall changes in elastic stiffness while elastic anisotropy can result from processing-induced,

preferred grain orientation and from alignment of aspheric pores or microcracks. Establishing structure-property relationships permits improved ultrasonic sensing of graphite microstructural changes that signal service-related degradation. The focus of this work is on ultrasonic approaches for sensing graphite microstructural changes that might occur as a result of service in nuclear reactors. In particular, porosity changes brought about by oxidation as well as anisotropy variations linked to neutron irradiation are of particular concern. In this work, ultrasonic characterization results for as-produced nuclear graphites are presented along with preliminary results for oxidized material. These results include measurements as a function of the ultrasonic mode (longitudinal and shear) and frequency. Characterization using these modes allows for determination of other moduli (assuming material isotropy) using a single measurement. In particular, results for Young's modulus as a function of material porosity are presented here. Additional measurements were made to assess effects related to propagation direction and polarization – these can be used to quantify material anisotropy. In particular, laser line sources have been used to quantify shear wave birefringence.

II. GRAPHITE MATERIALS

Commercially-produced, nuclear-grade graphites have complicated microstructures that relate directly to the manufacturing processes used to form them. Scanning electron micrographs of two types of graphite – IG-110 and PCEA – are shown in Fig. 1. IG-110 is fine grained with an average grain size of approximately 40 μm while PCEA has larger grains with an average size of 0.8 mm. In addition, processing affects the orientation distribution of the graphitic microstructural elements (IG-110 is isostatically pressed while PCEA is extruded). Both grain size and orientation affect damage accumulation from thermal cycling or neutron exposure [2], and extensive characterization of as-produced graphite is required to determine whether or not these types of service-related changes alter the properties sufficiently so that ultrasonic sensing of these changes can be successfully performed. Assessing structure-property relationships in various as-produced graphites as well as in materials that have undergone simulated, service-related changes will help in establishing the role that ultrasonics might serve.

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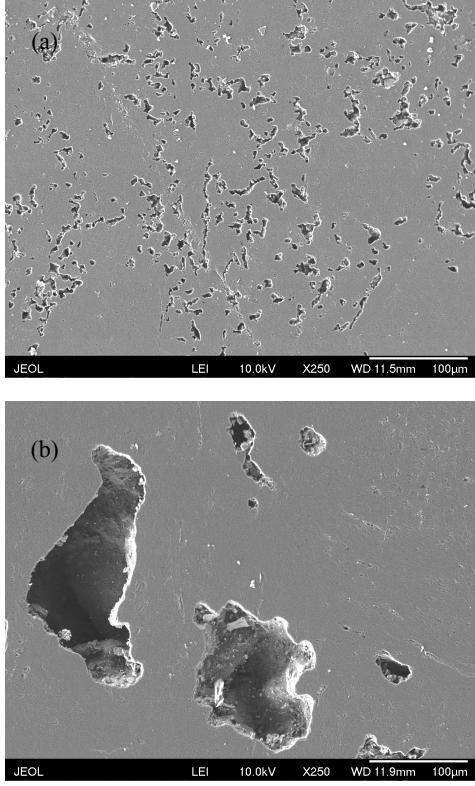


Fig. 1. SEM micrographs of (a) IG-110 and (b) PCEA showing pore distribution within the bulk of these materials.

III. EXPERIMENT AND RESULTS

In this work, elastic stiffnesses for 13 different currently-produced and legacy graphites have been studied and two of these have been oxidatively processed to alter pore volume and distribution. To measure modulus, laser ultrasonic measurements were made using a pulsed Nd:YAG laser source and detection was performed using optical interferometric methods based on Michelson-type instruments – see Fig. 2(a) for an experimental schematic. This source-receiver combination provides for non-contacting, highly linear transduction of broadbanded, ultrasonic pulses that allow for simultaneous determination of longitudinal and shear stiffnesses (if material density is known). Not only can these measurements be used to characterize materials as a function of the ultrasonic mode and frequency, but can also assess effects related to propagation direction if this is varied with respect to material frame of reference. In particular, shear wave polarization effects can be assessed by using laser line sources and can be used to quantify shear wave birefringence effects that depend wholly on material anisotropy and yield polarization-dependent shear stiffnesses.

A. Variation of Elastic Moduli with Graphite Density

By extracting ultrasonic times-of-flight from recorded waveforms (Fig. 2(b)) shows a representative waveform recorded for IG-110) along with measurements of sample dimensions and density, the longitudinal and shear moduli can be computed – results in Fig. 3(a) and (b) show these moduli as

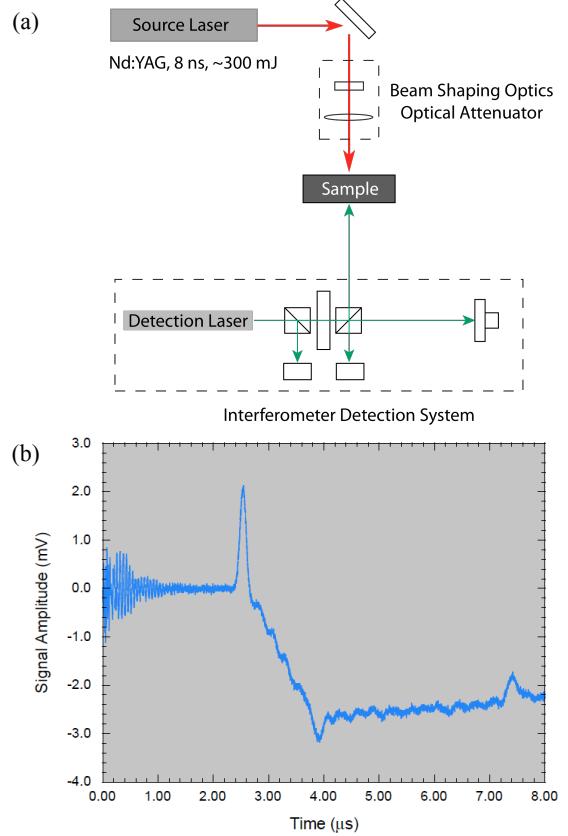


Fig. 2. (a) Experimental schematic of the apparatus used to make laser ultrasonic measurements on nuclear graphite samples. (b) Epicentral, laser ultrasonic waveform (A-scan) for IG-110 showing various ultrasonic arrivals that were identified to compute ultrasonic wavespeeds. Signals before the longitudinal wave arrival are related to electronic noise in the detection system.

a function of density along with a linear best fit to the data. Multiple samples of different orientations were made for each graphite. Scatter in the results shown in Fig. 3 can be attributed to material inhomogeneity and anisotropy. Despite the large variations in microstructure, the trend shows that higher densities are correlated with higher stiffnesses. Measurements show that among the graphites examined, a change in density of 0.21 g/cm^3 results in a change of 7.1 GPa for the longitudinal elastic stiffness and a change of 3.2 GPa for the shear stiffness.

B. Elastic Moduli vs. Porosity

To compare these results to others reported in the literature, these data were used to compute Young's modulus and porosity volume fraction. In Fig. 4, the variations of Young's modulus and the shear modulus with porosity volume fraction are shown. Young's modulus has been computed using the longitudinal and shear moduli and the volume fraction of porosity has been computed using the measured densities along with a theoretical density for graphite of 2.26 g/cm^3 . Linear fits to the values are shown and are extrapolated to zero porosity to provide an estimate of the theoretical stiffness of polycrystalline graphite based on these data sets – approximately 20.7 and 8.4 GPa for Young's modulus and the

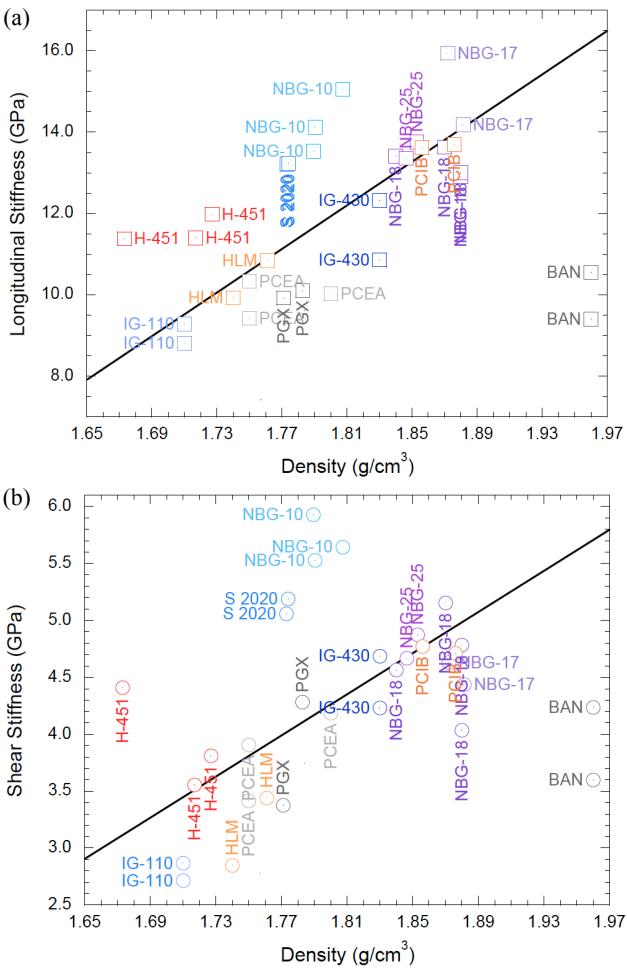


Fig. 3. Elastic modulus corresponding to (a) longitudinal material displacements and (b) shear displacement as a function of material density for different nuclear graphites (noted next to each data point). The solid lines represent best linear fits to the data (excluding points associated with BAN).

shear modulus respectively. These values are close to those computed by Kiewel *et al.* for graphite using the Reuss averaging method – 25.5 and 9.21 GPa [3]. However, the Reuss method is known to provide lower bounds for the elastic modulus in polycrystalline materials that are generally not observed – these results require additional consideration.

The assumed linear variation of elastic modulus with volume fraction is based on a model used by Shibata *et al.* to assess porosity effects on stiffness in two particular nuclear graphites, IG-110 and IG-430 [4]. At low volume fractions, this model predicts a linear variation of modulus with porosity. However, other variations can be assumed and will yield different values for the moduli of theoretically dense graphite. For example, early work by Cost *et al.* on a type of isotropic graphite assumed an exponential variation of modulus with porosity based on prevailing experimental evidence at that time [5]. Using an exponential fit to the data shown here yields modulus values at zero porosity of 32 and 13 GPa for Young's and the shear modulus respectively. While significantly higher than the Reuss values, these are still near the lower bounds

(39.9 and 14.9 GPa) for graphite provided by Kiewel *et al.* [3] based on theory by Hashin and Shtrikman [6,7] that assumes random grain orientation. Another variation developed by Wagh *et al.* suggests a power law relationship between modulus and density – $E \propto \rho^n$ where E is Young's modulus, ρ is the material density and n is an exponent typically between 2 and 3 for a variety of measurements on porous materials [8]. A power law fit to the data presented here yields exponents much higher indicating that the modulus varies more strongly with porosity than predicted by this model.

Models that consider only the effect of porosity on modulus do not appear to correctly predict the variations presented here. The stiffnesses of these graphites are far lower than might be expected if porosity alone were solely responsible for modulus variations and other microstructural elements likely contribute. The effect of microcracking on modulus needs to be considered since microcracking occurs in graphite and is known to lower material moduli. The results here support the suggestion that microcracking plays a significant role in determining graphite stiffness since theoretically dense graphite containing microcracks would have stiffnesses much lower than those predicted using models based on single crystal stiffness values. Also, assuming microcrack density varies with porosity, the variations of moduli with density could surpass those expected for materials that contained only porosity. Being able to separate the effects of porosity on modulus from those related to microcracking could be important in interpreting service-related changes that occur in various nuclear graphites.

To simulate one type of service-related condition, high temperature oxidation of IG-110 and NBG-18 was performed – only results for IG-110 will be mentioned here. Oxidation primarily affects graphite by decreasing material density (increasing the volume fraction of porosity) [9]. The overall effect of this type of process is to decrease material stiffness which can have significant consequences for the performance of the material. Preliminary oxidation of IG-110 has shown this effect. Heating a sample of IG-110 in a flowing oxygen-containing atmosphere at 500 °C until it reached a nominal

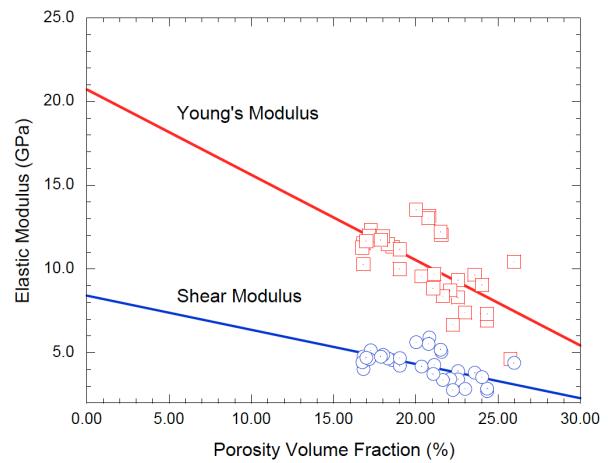


Fig. 4. Graphite modulus as a function of porosity volume fraction shown with linear fits to the data for Young's modulus and the shear modulus.

weight loss of 10% produced a material with longitudinal and shear moduli of 5.14 and 1.93 GPa respectively and a density of 1.68 g/cm³. These values fall below the trends established by the best fit lines shown in Fig. 3 – this might be related to a breakdown of porosity-based models for the stiffness of this oxidized material.

C. Shear Birefringence Measurements

Beyond influencing the overall stiffnesses of materials, microcracks can also affect material isotropy. Some graphites, such as IG-110, are processed using isostatic pressing that should result in materials that are highly isotropic. Others, including PCEA and NBG-10, are extruded and this type of processing can induce significant anisotropy related to preferred grain orientation and aligned microcracking. The associated effects on the elastic properties can be measured using ultrasonic, shear wave, birefringence measurements.

For the results presented here, shown in Fig. 5, a laser ultrasonic line source has been used to produce linearly polarized shear waves. The polarization of these waves can be adjusted by rotating the line source relative to the sample. The isotropy of the material can be measured by recording ultrasonic waveforms as a function of the rotation angle and noting changes in the times-of-flight for various ultrasonic modes. Results in Fig. 5(b) clearly show the isotropy of IG-110 – the A-scan waveforms are nearly identical for all line source orientations – while the shear wavespeed in PCEA varies considerably with line source orientation (see Fig. 5(c)). The shear wave time-of-flight varies by approximately 8% for the results shown while the longitudinal wavespeed remains constant.

IV. CONCLUSIONS

This work has shown aspects of laser ultrasonic characterization of nuclear graphites and has highlighted aspects of the structure-property relationship that must be studied in more depth to determine the utility of laser ultrasonics for characterization of service-related changes that are known to occur in these materials.

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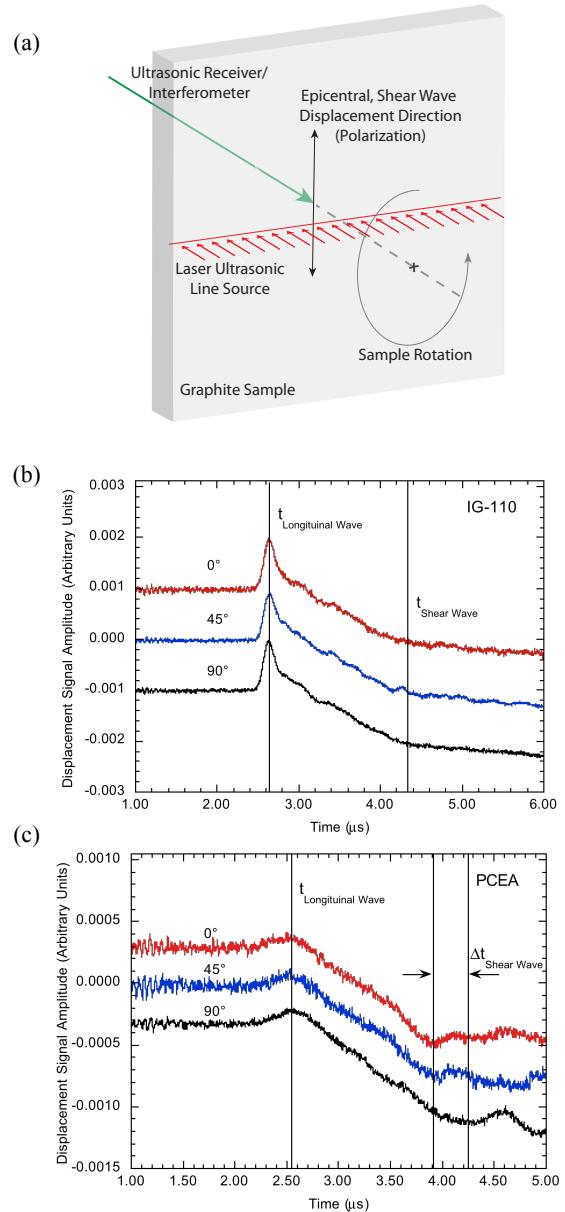


Fig. 5. (a) Diagram of laser ultrasonic line source measurement geometry used in this work to assess shear wave birefringence effects. Laser ultrasonic line source measurements for various line orientations on (b) IG-110 and (c) PCEA. Longitudinal and shear wave arrivals are noted. No shear birefringence in IG-110 indicates this material is at least transversely isotropic while PCEA has significant variations in the arrival times of shear waves indicating anisotropy in this material.