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MECHANICAL PROPERTIES OF NUCLEAR FUEL SURROGATES USING PICOSECOND LASER ULTRASONICS

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ABSTRACT

Detailed understanding between microstructure evolution and mechanical properties is important for designing new high burnup nuclear fuels. In this presentation we discuss the use of picosecond ultrasonics to measure localize changes in mechanical properties of fuel surrogates. We develop measurement techniques that can be applied to investigate heterogeneous elastic properties caused by localize changes in chemistry, grain microstructure caused by recrystallization, and mechanical properties of small samples prepared using focused ion beam sample preparation. Emphasis is placed on understanding the relationship between microstructure and mechanical properties.

INTRODUCTION

Microstructure mediated mechanical properties of nuclear fuel are closely related to fuel longevity and reactor safety. While in operation, nuclear fuel microstructure undergoes a tremendous transformation brought about by neutron irradiation. Changes include void and bubble formation, changes in dislocation density, grain restructuring due to recrystallization and changes in composition due to fuel cladding chemical interaction. Complicating this picture is the fact that mechanical properties change dramatically from the fuel pin center to the fuel pin exterior. Thus there is a need to develop new characterization techniques that can provide spatially resolved information regarding mechanical properties. In this presentation we explore using picoseconds ultrasonics to measure localized elastic properties of metallic and ceramic fuel surrogates.

EXPERIMENT

The experimental setup used for generating and imaging ultrasonic waves is shown in Fig. 1. The pump and probe beams, with wavelengths of 400 and 800 nm are derived from a Ti:sapphire laser

with a pulse duration of ~200 fs. The ultrasonic imaging system is similar to the setup presented by Tachizaki et al.[1] and involves translation of a probe beam relative to a fixed pump beam. The experimental apparatus can employ either a single probe pulse for monitoring changes in probe reflectivity or two interferometric pulses for monitoring changes in optical phase and reflectivity. The probe is then sent through a telescope and focused onto the sample using a 50x microscope objective. Both lenses of the telescope have a 100 mm focal length. The first lens is attached to a stage system that allows the lens and the probe beam to be translated in the x-y plane (the probe beam propagation vector remains collinear with the lens axis). The second lens serves to converts the x-y motion of the beam into a change in entrance angle into the objective. Since the objective is placed at the focal point of the second lens, the entrance angle can be changed without translation in the x-y plane (a necessity for microscope objectives with small entrance apertures). The pump beam is guided along the optical axis of the objective using a dichroic beam splitter placed after the telescope. The spot diameter of both beams at the sample is ~ 2μm at full width at half maximum intensity. This system provides a convenient means to scan the probe beam relative to the pump beam for situations that allow for only single-sided access.

For this study we use highly textured copper as a surrogate for metallic fuel and depleted uranium oxide as a surrogate for ceramic fuel. Acoustic waves are excited thermoelastically in our metallic sample. For our ceramic sample, which is a semiconductor, both the thermoelastic and deformation potential mechanisms are involved in acoustic generation.

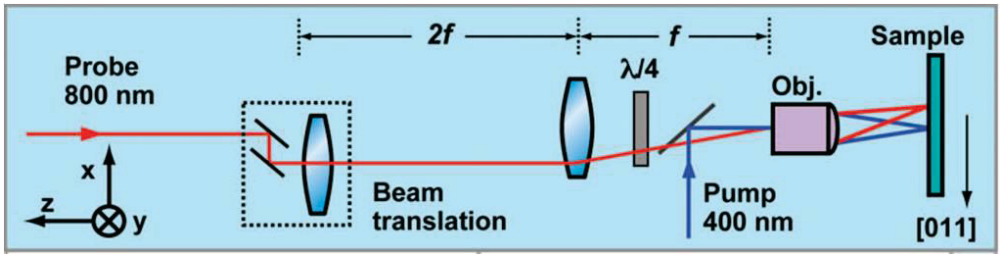

Figure 1. Experimental seup. Both pump and probe are focus through a single microscope objective.

RESULTS AND DISCUSSION

Here we explore three distinct lines of investigation. The first involves using high frequency surface acoustic waves to measure mechanical texture caused by deformation and recrystallization. The second entails performing picoseconds ultrasonic experiments on thin films fabricated from bulk samples using focused ion beam (FIB) fabrication techniques. Here the spatial resolution is defined by the sample size. Lastly, we examine using Brillouin oscillations in uranium oxide as a method to monitor defects caused by charged particle irradiation.

Surface Acoustic waves

Metallic fuel can develop mechanical texture either through plastic deformation during processing or due to the production of small irradiation induced dislocation loops during operation.[2] For materials that have strong single crystal elastic anisotropy, texture can profoundly influence polycrystal elastic properties. In this section we use high frequency surface acoustic waves to

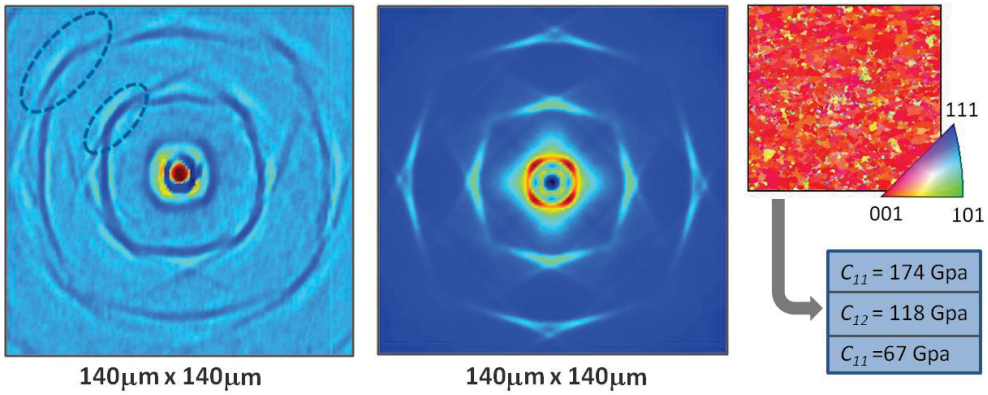

Figure 2. Left pane: An interferometric image of surface acoustic waves propagating on a highly textured copper sample. Middle pane: A model of surface acoustic waves using an angular spectrum of plane waves. The elastic constants used for this model were obtained from the EBSD data shown in the right pane.

measure elastic anisotropy caused by texture. A textured microstructure was imparted by rolling a high purity copper sample. A reduction of 97.5% was obtained by rolling the material many times. Upon annealing, a strong texture develops that is almost completely cubically aligned (here the crystallographic axes align with the surface normal and the rolling direction).[3] An electron backscatter diffraction (EBDS) micrograph, showing the orientation of the crystallites, is presented in the right pane of Fig. 2. A typical image of surface acoustic waves in this sample is shown in the left pane of Fig. 2. The concentric wave fronts, due to the 76 MHz periodic excitation, exhibit distinctive phonon focusing due to elastic anisotropy. The middle image shows a model of surface acoustic waves in this sample. The model accounts for acoustic diffraction from a point source by using an angular spectrum of plane waves. The elastic constants used for this model were obtained by forming a polycrystalline average elastic stiffness tensor using the EBSD data.[4] The pseudo surface acoustic wave, which exists for this particular sample,[5] was not modeled. In the experimental data shown in the left pane of Fig. 2, the pseudo surface wave is easily observed and is denoted by the dashed ovals. The reasonable agreement between model and experimental data suggests a new method for localized measurement of mechanical texture.

Picosecond Acoustics on FIB sample

In this section we explore applying picosecond ultrasonics to study elastic properties of bulk materials. Traditionally picosecond ultrasonic techniques employee longitudinal bulk waves to investigate the dimensional and elastic properties of thin films. This is because the acoustic echo emanating from the film/substrate surface is easily observed in a relatively short time window. For samples thicker than a few tens of microns, observing the acoustic echo is difficult for two related reasons: 1 - the amplitude of the acoustic echo diminishes due to scattering and geometric attenuation, 2 - finding the acoustic echo in a specific time window becomes a demanding task due to uncertainty in sample thickness. For these reasons picosecond ultrasonics is not typically applied to study the elastic properties of thick samples. Addressing this difficulty, we used picoseconds acoustic to measure elastic properties of specially prepared FIB samples. Typically FIB fabrication techniques are used to provide electron transparent samples for TEM analysis. Here we use an approach, similar to the lift-out technique, to provide a simply supported thin film. This approach essentially involves locating an area of interest and removing a small sliver for

picosecond acoustic measurements (illustrated in the left pane of Fig. 3). We used a high purity polycrystal copper sample for this proof of principle experiment. The sample dimensions are approximately 10 x 10 μm and the thickness is approximately 350 nm. A typical waveform using interferometric detection is shown on the left pane of Fig. 3. The broad echoes are due to ultrafast electron diffusion and are consistent with a previous study.[6] Future work will involve using this approach to relate elastic properties to chemical composition in fuel/cladding diffusion bonded samples.

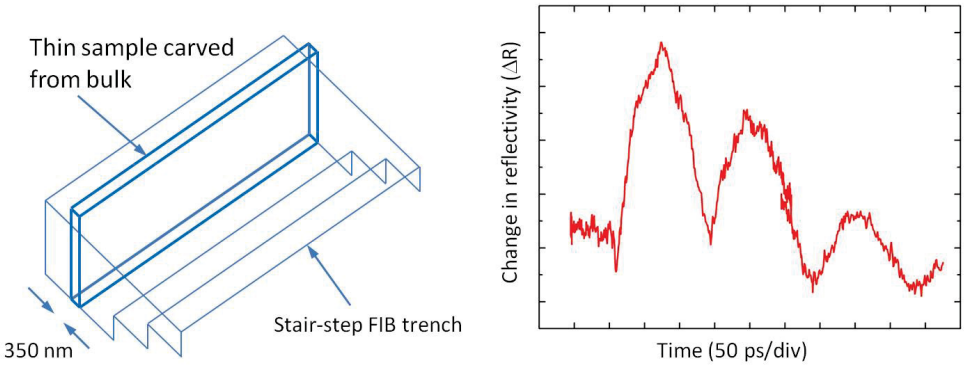

Figure 3. Picosecond ultrasonic echoes in a FIB prepared sample. The sample fabrication technique is illustrated in the left pane and a typical waveform is shown in the right pane.

Brillouin Oscillations in Uranium Oxide

In this section we investigate the utility of picosecond ultrasonics to characterize damage in proton irradiated uranium oxide. Two uranium oxide single crystal samples having the same orientation were studied. One is un-irradiated and the other has been irradiated with 2.5 MeV protons to a dose of 0.4 displacements per atom at 800C. An ion transport simulation suggests that the damage layer extends to a depth of approximately 25 μm. A typical picosecond ultrasonic signal for both samples is shown in Fig. 4. The pump pulse arrival is followed by a high frequency Brillouin oscillation at approximately 28 GHz that arises from interference between two portions of the reflected probe pulse,[7] part reflected from the surface, and part from a strain-induced change in the refractive index associated with the bulk longitudinal ultrasonic pulse in the uranium oxide sample. For a normally incident probe pulse, the period of this oscillation is given by $TL=\lambda/2nV_L$, where λ is the probe optical wavelength, n is the probe index of refraction (2.12), and V_L is the velocity of the longitudinal acoustic phonons (5670 m s^{-1}).[8] The damping of this mode is primarily governed by the optical attenuation of the probe light in the uranium oxide sample. The corresponding Fourier amplitude spectrum is shown on the right pane of Fig. 4.

The Brillouin oscillation of the un-irradiated sample exhibits little attenuation and is centered at ~27 GHz. In contrast the oscillations in the irradiated sample decay rapidly and the center frequency is slightly shifted. The broadening and shift in the frequency spectrum is most likely due to a coupling between irradiation damage and the index of refraction. It is well know that irradiation introduces a high concentration of point defects which will influence the real and

imaginary parts of the index of refraction. It is envisioned that this approach could be used to look at the evolution of point defects in nuclear fuel samples during thermal cycling. Understanding the defect evolution would provide important validation metrics for fundamental models of materials in extreme irradiation environments.

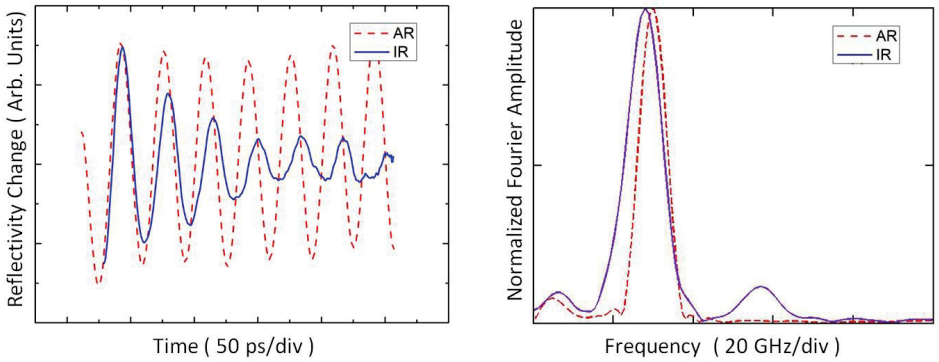

Figure 4. Brillouin oscillations in uranium oxide. The oscillations in the irradiated sample exhibit a rapid decay. The Fourier amplitude spectrum for both samples is shown in the right pane.

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[8] The longitudinal velocity is a function of orientation. Currently we do not know the exact orientation of the sample but for the sake of discussion we have assumed $V_L=(C_{11}/\rho)^{1/2}$.