Enabling TRISO Metrology Using Mueller Matrix Imaging

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Abstract

Tristructural-isotropic (TRISO) fuel particles represent a leading advancement in nuclear fuel technology for high-temperature gas-cooled reactors (HTGRs) and emerging Generation IV systems. Each particle consists of a uranium, thorium, or mixed-oxide fuel kernel encapsulated by three concentric layers: porous carbon, inner pyrolytic carbon, and silicon carbide, followed by an outer pyrolytic carbon coating. This multilayer architecture provides exceptional fission product retention, structural integrity under irradiation, and resistance to high temperatures, often exceeding 1600°C. The porous buffer layer accommodates fission gases and swelling, while the SiC layer serves as a miniature pressure vessel, providing both mechanical strength and an effective diffusion barrier against metallic fission products. Collectively, these features mitigate the risk of catastrophic fuel failure, a primary safety concern in conventional light water reactor fuels. However, if the particle shell is damaged, it creates a huge safety concern. Mueller Matrix Imaging (MMI) enables researchers to learn about the structural integrity of the TRISO particles, ensuring their safety for use.

Introduction

Mueller Matrix Imaging is a new form of metrology that allows precision assessment of the polarization properties of a sample. This is especially important for learning about the structural integrity of TRISO particles and can determine if the particles in a batch are deformed and/or have appropriately low anisotropy of the deposited graphite layers.

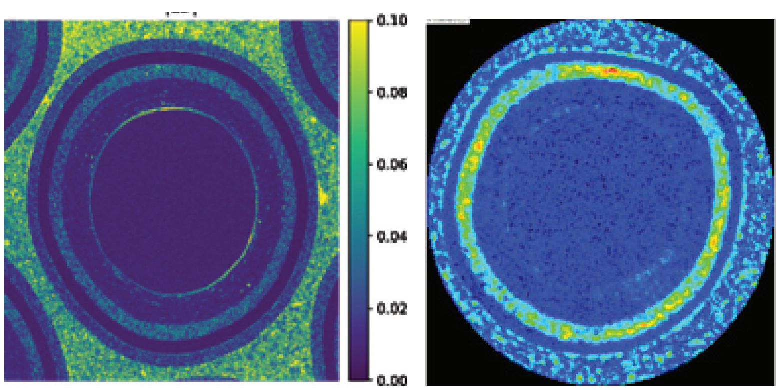

Figure 1: Side-by-side comparison of linear diattenuation in TRISO particle using the Mueller Imager (left) and 2MGEM (right). [6]

The first TRISO characterization attempts headed by Gerald E. Jellison involved a system called the “Two Modular Generalized Ellipsometry Microscope” or 2MGEM for short. This characterization technique involved sending 560nm sodium line light through a photoelastic modulator (PEM), which is then focused on the sample and reflected back through a second PEM. This signal is then separated by a beam splitter and read by both a camera for sample alignment and a photomultiplier tube (PMT) for measurement of the sample's Mueller matrix.

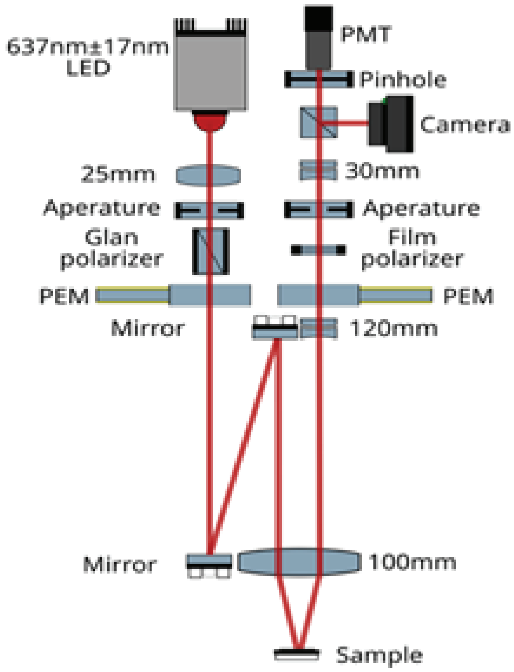

Figure 2: Optical path of Two Modulator Generalized Ellipsometry Microscope.

While this technique is effective, scanning across the sample is slow compared to whole field imaging. In the case of TRISO imaging, production batch sizes involve thousands or millions of particles, and measuring representative samples requires numerous measurements. Mueller Matrix Imaging increases the speed of the characterization process.

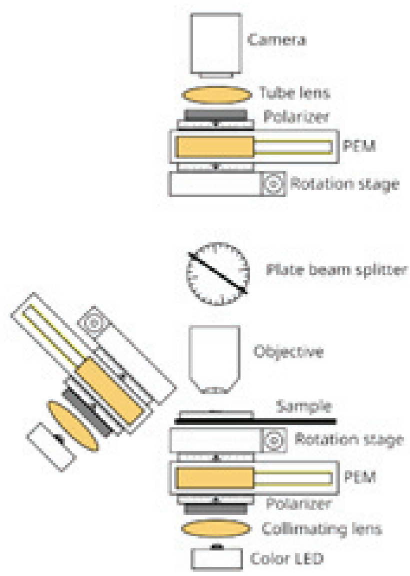

Figure 3: Sketch of Mueller Matrix Imager with reflection arm at 36°.

The Mueller Matrix Imager simplifies the optical path of reflection-based imaging by eliminating the PMT and adds the capability to measure both reflection and transmission properties. Unlike the 2MGEM system, which collects data point-by-point and requires up to 30 minutes to acquire a 256×256 datapoint image over a 1 mm pellet, the Mueller Imager captures its dataset in a fraction of the time. Each element of the Mueller matrix requires only two frames and

a DC intensity image for a given PEM orientation, resulting in a total of 33 frames (3 DC, 15 positive, 15 negative) acquired in roughly 3.3 seconds. The data is collected using the pco.edge 5.5 USB sCMOS camera by Excelitas, which has a pixel resolution of 2560 $\times$ 2160 pixels.
The original 2MGEM system can measure the full Mueller matrix at 560 nm (the sodium line), but for TRISO samples it typically measures only nine of the matrix elements. Capturing all 16 elements would require three full scans, approximately one hour of total acquisition time. By comparison, our in-house Mueller imaging system measures nine elements at 633 nm and can achieve equivalent measurements in just 0.5 seconds. Even for a full Mueller matrix scan, our system would require only about 6 seconds, compared to roughly an hour on the 2MGEM, highlighting its substantial advantage in speed and experimental efficiency.

Methods

TRISO particle samples were provided by Oak Ridge National Laboratory (ORNL) for imaging development and evaluation using the Mueller Matrix Imager. Each TRISO sample puck contained an average of 37 fuel pellets, arranged to provide a representative distribution of particle types and orientations.
All measurements were conducted using the reflection arm of the Mueller Imager, which is designed to characterize polarization properties from reflected light rather than transmitted light.
During data acquisition, the system captured a complete set of Mueller matrix elements for each region of interest using sequential modulation and demodulation of the polarization states. The reflection-based setup enabled precise measurement of polarization-dependent signatures across the surface of each TRISO pellet, supporting subsequent analysis of particle uniformity and defect detection performance.

Experiment:

Imaging experiments were conducted using the reflection arm of the Mueller Matrix Imager to characterize TRISO fuel particles provided by Oak Ridge National Laboratory. Each TRISO sample puck contained approximately 37 fuel pellets, arranged to ensure representative coverage of particle morphology and surface characteristics.

During each experiment, the reflection geometry was used to measure polarization-dependent properties from the pellet surfaces. The system captured a complete set of Mueller matrix elements by sequentially modulating and analyzing the polarization states of the incident and reflected light. Each dataset consisted of 33 total frames: 3 DC intensity images, 15 positive, and 15 negative polarization states. The total exposure time for the full dataset was approximately 3.3 seconds, with an overall acquisition and processing time of roughly 60 seconds per field of view. Images were analyzed using in-house software to reconstruct the Mueller matrix at each pixel, enabling visualization of polarization parameters such as diattenuation, retardance, and depolarization across the TRISO surfaces. These measurements provided spatially resolved information about particle uniformity, surface defects, and internal stress patterns, supporting the development of rapid imaging techniques for TRISO fuel characterization.

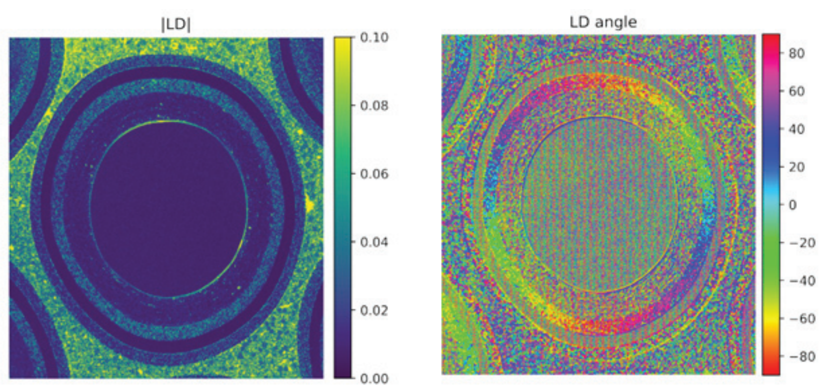

Figure 4: Linear diattenuation and linear diattenuation angle of TRISO particle.

Emphasis was placed on linear diattenuation (LD) and the linear diattenuation angle (LDA), which describe the differential absorption of light polarized along two orthogonal axes and the orientation of that anisotropy, respectively. In TRISO fuel particles, variations in LD can reveal directional differences in surface microstructure, carbon layer anisotropy, or residual stress from coating processes. The LDA provides complementary insight into the alignment or preferred direction of these anisotropic features. Together, LD and LDA serve as sensitive indicators of material uniformity and process consistency, allowing the Mueller Matrix Imager to detect subtle deviations in particle structure that may influence mechanical strength, optical response, or fuel performance.

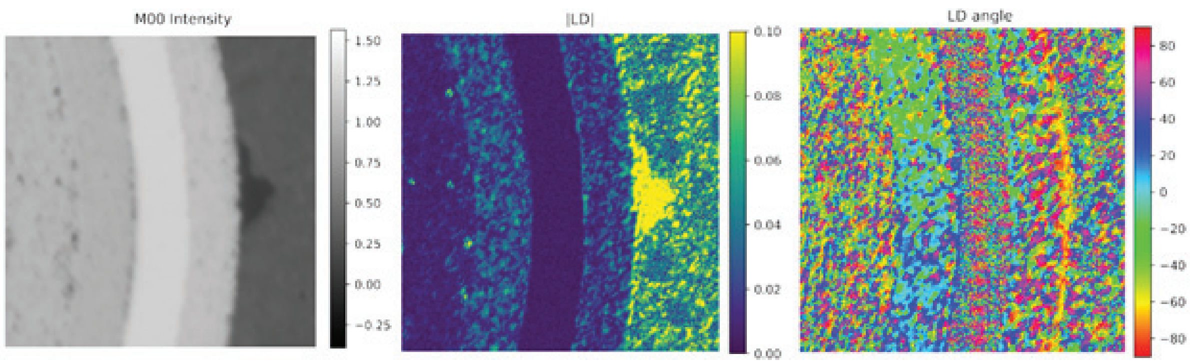

Figure 5: Different outputs and how they each pick up on surface deformation of the TRISO particle.

The quantitative relationships used to derive these polarization properties are defined by the following equations:

$$|N| = \left| \frac{R_o - R_e}{R_o + R_e} \right|$$

$$CD = \frac{R_l - R_r}{R_l + R_r}$$

R_o and R_e represent the reflected intensities of light polarized along the orthogonal axes of the sample's birefringent structure, defining the normalized linear diattenuation magnitude /*N*/. Similarly, R_l and R_r correspond to left- and right-circularly polarized reflection intensities, defining the circular diattenuation CD. These quantities provide a basis for distinguishing polarization-dependent absorption effects in the TRISO coating layers. The LD and CD maps generated from these expressions help identify anisotropic regions, revealing differences in optical path and structural uniformity that correlate with coating integrity and potential performance variations in the fuel particles.

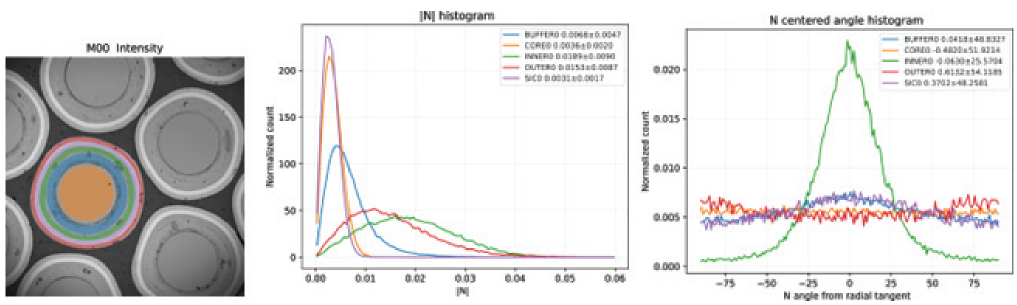

Figure 6: Color analysis of TRISO particles was used to create histograms of the layer quality.

Discussion

The results of these experiments demonstrate that the Mueller Matrix Imager provides a precise and efficient, method for analyzing the optical anisotropy of TRISO fuel particles. By using the reflection arm of the system, we were able to obtain high-resolution polarization data directly from the pellet surfaces without requiring transmission through the sample. This configuration is particularly valuable for opaque or layered materials such as TRISO coatings, where internal structures influence reflected polarization states.

The rapid data acquisition, on the order of seconds rather than minutes, enables practical, large-scale imaging of entire sample pucks containing dozens of fuel particles. This capability significantly enhances throughput for quality assurance or process monitoring, allowing for statistical evaluation of coating uniformity across multiple particles. The reconstructed Mueller matrices revealed consistent polarization patterns associated with the layered TRISO architecture, while local deviations in diattenuation and retardance suggested the presence of surface irregularities or stress-induced anisotropy.

Overall, the Mueller Matrix Imager's ability to rapidly quantify and visualize polarization-dependent features in TRISO fuel particles underscores its potential as a diagnostic and process-development tool. Future work will focus on correlating the observed LD and LDA distributions with known manufacturing variables and destructive verification methods, enabling the establishment of optical benchmarks for TRISO quality assessment. This advancement represents a meaningful step toward implementing high-speed polarization imaging as a standard method for fuel particle evaluation.

References

1. Jellison, G. E., and D. H. Lowndes. "Time-Resolved Ellipsometry Measurements of the Optical Properties of Silicon during Pulsed Excimer Laser Irradiation." Applied Physics Letters, vol. 47, no. 7, 1 Oct. 1985, pp. 718–721. AIP Publishing, https://doi.org/10.1063/1.96014
2. Jellison, G. E., and F. A. Modine. "Two-Modulator Generalized Ellipsometry: Theory." Applied Optics, vol. 36, no. 31, 1 Nov. 1997, p. 8190. Optica Publishing Group, https://doi.org/10.1364/ao.36.008190
3. Jellison, G. E., G. L. Petersen, et al. "Nuclear Quadrupole Resonance Studies of Amorphous, Orthorhombic, and Rhombohedral Arsenic." Physical Review B, vol. 22, no. 8, 15 Oct. 1980, pp. 3903–3916. American Physical Society, https://doi.org/10.1103/physrevb.22.3903
4. Jellison, G. E., D. E. Holcomb, et al. "Internal Electric Field Measurements in Cadmium Zinc Telluride Using Transmission Two-Modulator Generalized Ellipsometry." Journal of Electronic Materials, vol. 32, no. 7, July 2003, pp. 789–795. Springer, https://doi.org/10.1007/s11664-003-0072-8
5. Jellison, G. E. "Generalized Ellipsometry for Materials Characterization." Thin Solid Films, vol. 450, no. 1, Feb. 2004, pp. 42–50. Elsevier, https://doi.org/10.1016/j.tsf.2003.10.148
6. Jellison, G. E., J. D. Hunn, and A. H. Breninger. Upgrade and Operation of the Second Generation Two-Modulator Generalized Ellipsometry Microscope 2-MGEM2. Oak Ridge National Laboratory, Apr. 2013.
7. Jellison, G. E., T. Aytug, et al. "Optical Properties of a Nanostructured Glass-Based Film Using Spectroscopic Ellipsometry." Thin Solid Films, vol. 617, Oct. 2016, pp. 38–43. Elsevier, https://doi.org/10.1016/j.tsf.2015.12.046
8. Jellison, G. E., D. N. Leonard, L. M. Anovitz, C. M. Parish, et al. "Crystallographic Orientation of Uniaxial Calcite and Dolomite Determined Using Reflection Generalized Ellipsometry." Journal of Applied Physics, vol. 124, no. 22, 14 Dec. 2018. AIP Publishing, https://doi.org/10.1063/1.5048724
9. Jellison, G. E., D. N. Leonard, L. M. Anovitz, M. C. Cheshire, et al. "Crystallographic Orientation of Orthorhombic Aragonite Using Reflection Generalized Ellipsometry." Journal of Applied Physics, vol. 126, no. 4, 23 July 2019. AIP Publishing, https://doi.org/10.1063/1.5109093

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