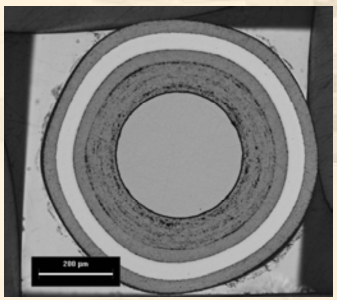
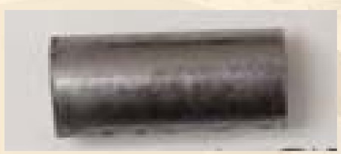
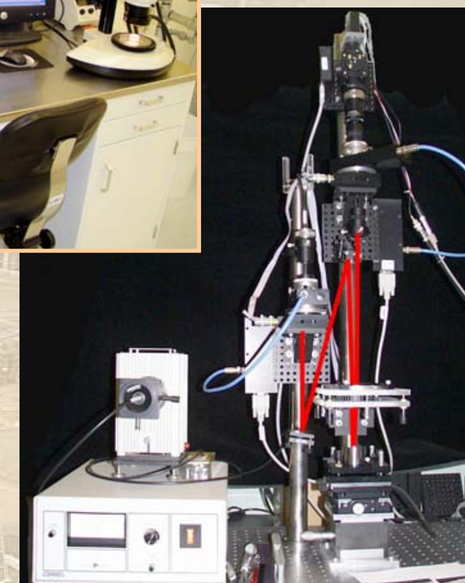
UT-BATTELLE

Advanced characterization methods for TRISO fuels







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Purpose

- Provide overview of TRISO particle fuel characterization effort at ORNL.

- Describe new and modified methods developed at ORNL for analyzing kernels, coated particle fuel, and compacts.

  Computer automated microscopy

  Density column measurements

  Pyrocarbon anisotropy measurements

  Impurity leaching from compacts

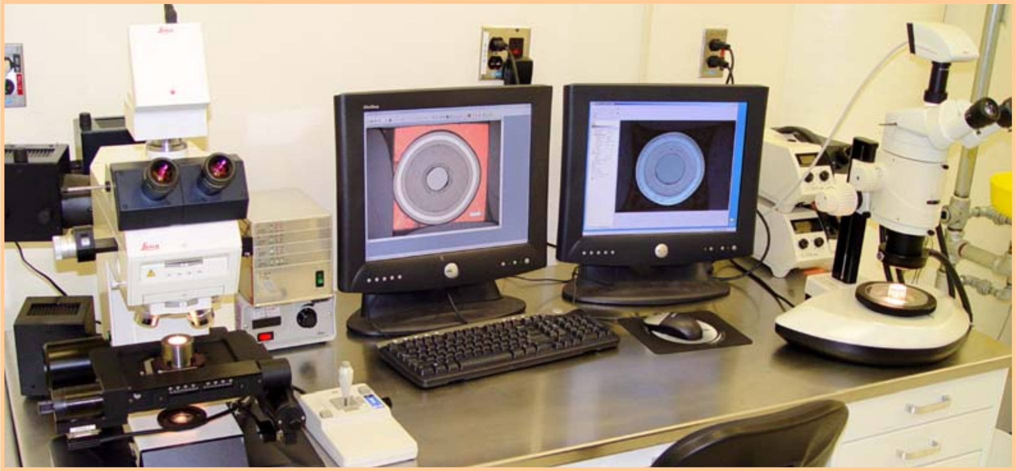
ORNL particle fuel research

Ongoing research in the areas of:

- kernel production
- coating development
- compact production

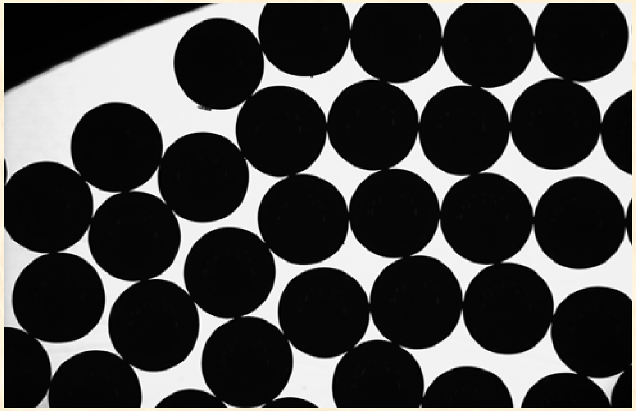
Characterization methods are being developed for kernels, coated particles, & compacts.

Computer automated microscopy



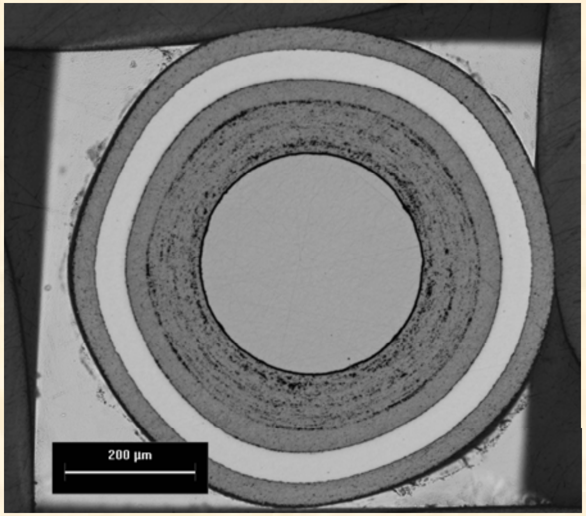
- computer-driven stage
- digital image acquisition
- image analysis software written in-house

SHADOW IMAGES



measure size & shape of whole kernels, coated particles, and overcoated particles

CROSS-SECTIONS



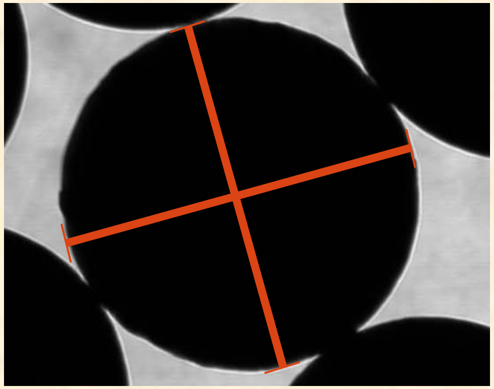
measure layer thicknesses of coated particles

Shadow images for size & shape

Conventional technique:

"Manual" measurement of average particle size, maximum diameter, and minimum diameter (or perpendicular diameter).

- operator-dependent measurement error
- small sample size (~50-100)

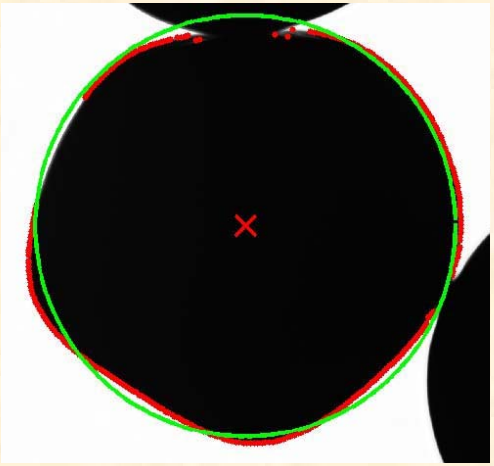


Particles sometimes are still measured with a reticule micrometer.

ORNL technique:

Computer identification of particle center, radial measurements of particle edge, Fourier transform solution of particle boundary.

- measurement error depends on optical resolution
- large sample sizes (~2000-5000)



Particle image was extracted and analyzed by computer.

Key advantages

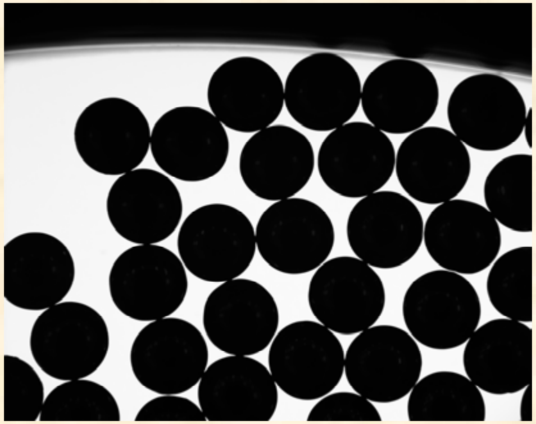
Data acquisition is easier and more consistent.

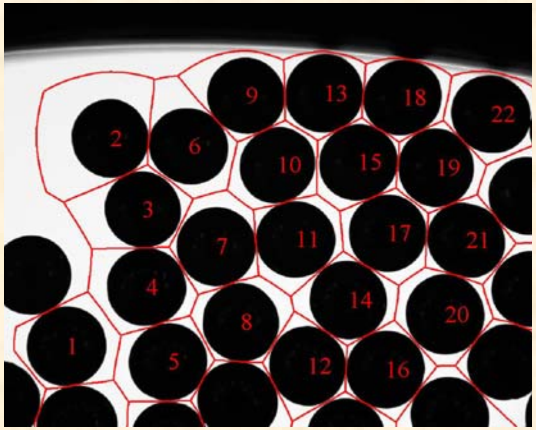
Measurements are typically more accurate.

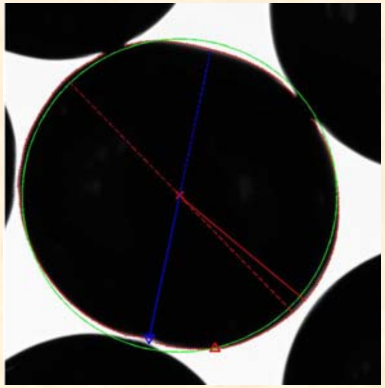
Statistical analysis greatly benefits from ~100x increase in sample size.

Equation for particle boundary could be used in fuel performance models.

New metrics for particle shape have been developed that should be more closely linked to fuel performance.







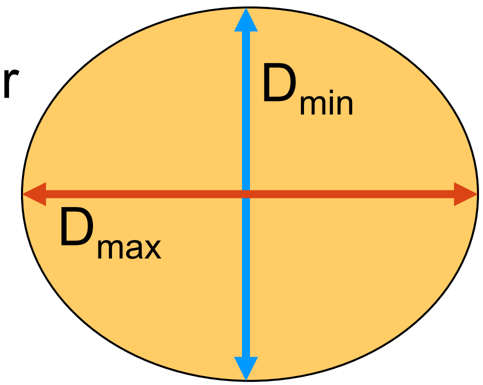
Metrics for particle shape

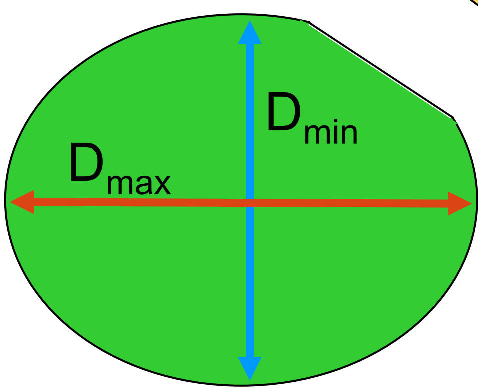
CURRENTLY USED

Diameter Aspect Ratio

$$\frac{D_{max}}{D_{min}}$$

Great metric for symmetric aspherical shapes.





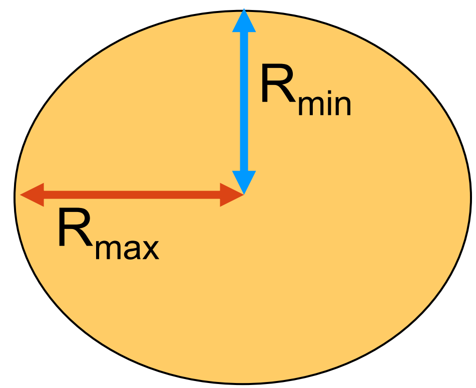
Diameter ratio is less sensitive to asymmetric features.

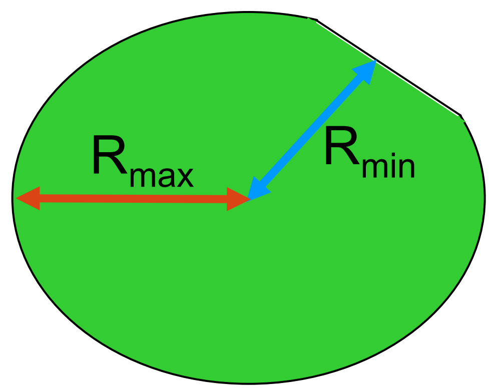
Radius Aspect Ratio

$$\frac{R_{max}}{R_{min}}$$

Identification of particle center by computer allows easy calculation of radius ratio.

Radius ratio is also great for symmetric aspherical shapes.



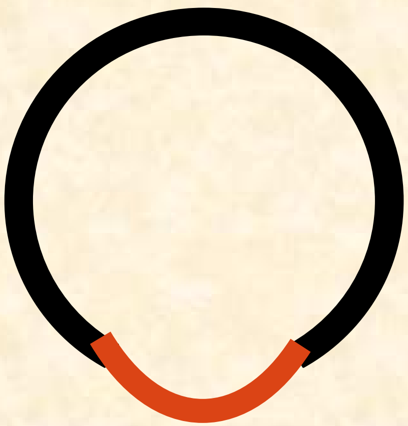


Radius ratio is more sensitive to asymmetric features.

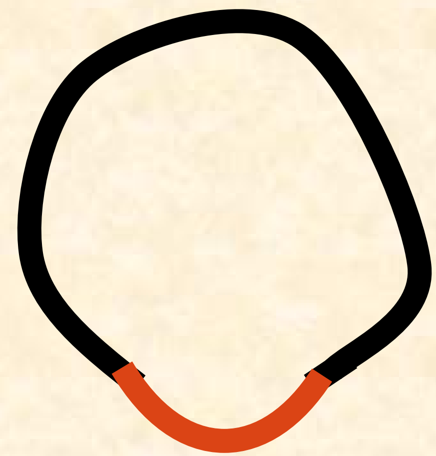
New metric for shape

The mechanical strength of layers is related to localized shape features of kernels and coated particles.

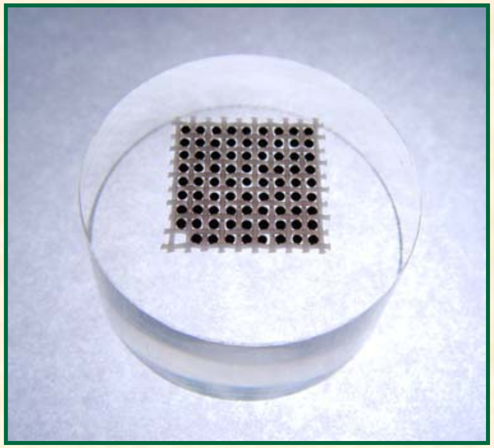
$R\kappa_{max}$ **the product of the radius and curvature at the point of maximum curvature.**



Local stress primarily depends on the local shape, not the overall shape features.

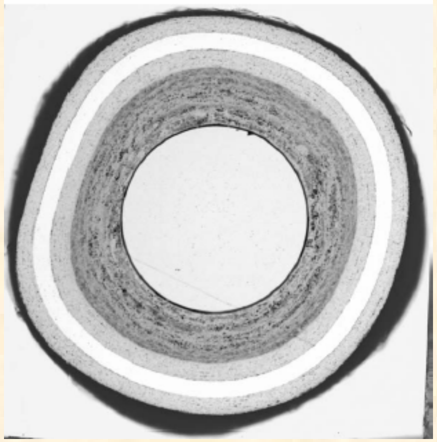


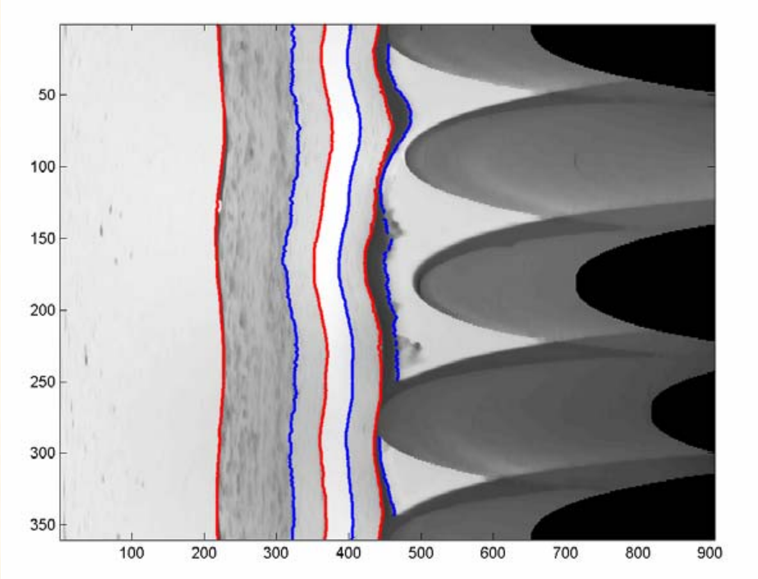
Layer Thickness Measurements



Particles mounted and polished in a grid.

Automatic image acquisition based on the grid mesh size.





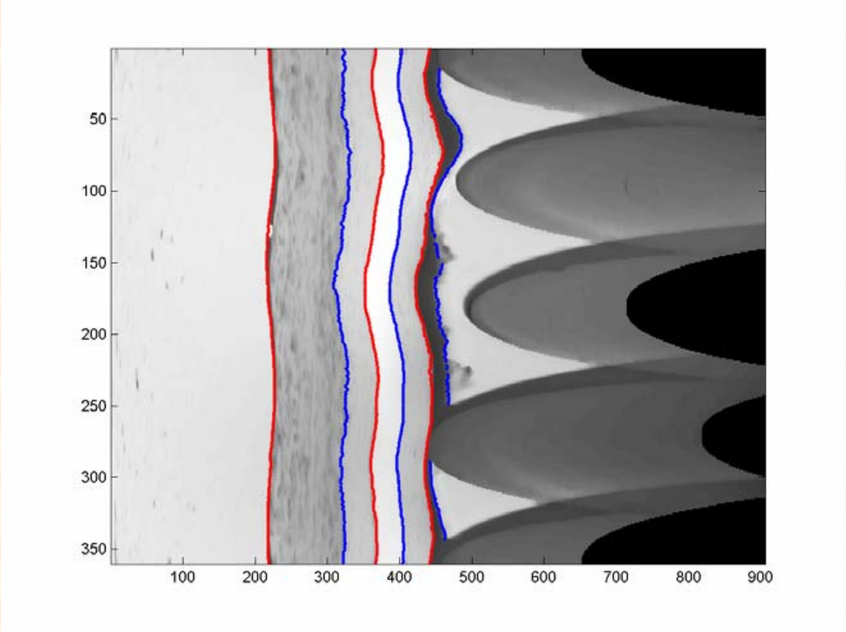
Images are radially “unwrapped.” Layer boundaries are measured at 1° intervals.

Benefits

Computer automated optical microscopy is commonly done for 99 particle mounts.

practical sample size of ~100-300 particles

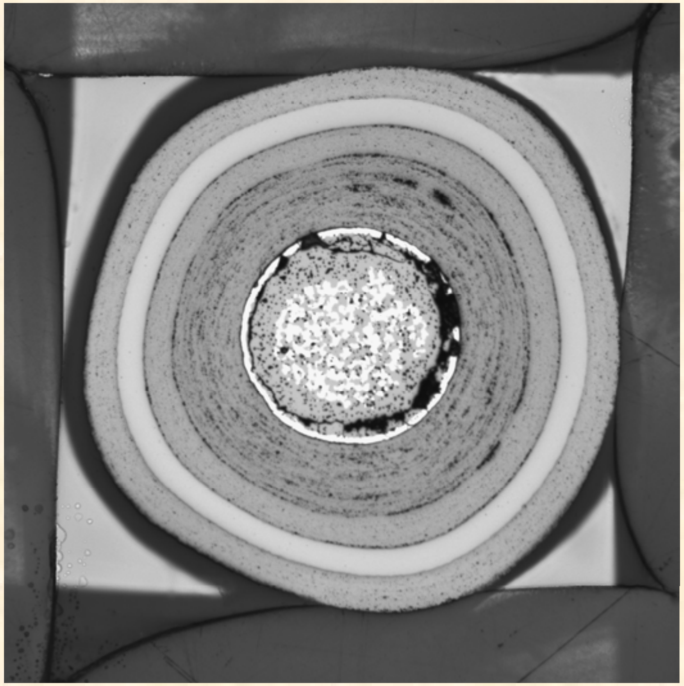
Measured layer thicknesses of <u>each</u> particle are based on hundreds of data points.



No operator bias in identifying boundaries or selecting measurement locations.

Off-midplane correction of measurements can be readily applied.

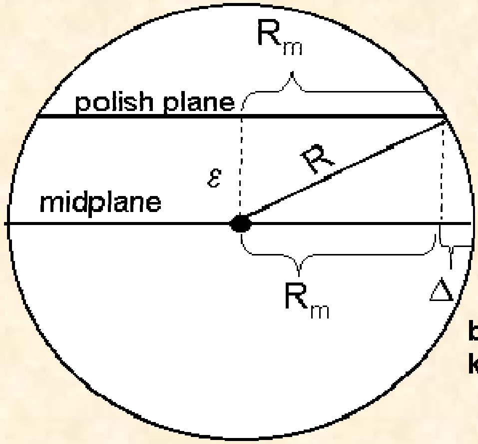
Layer thicknesses from cross-sections

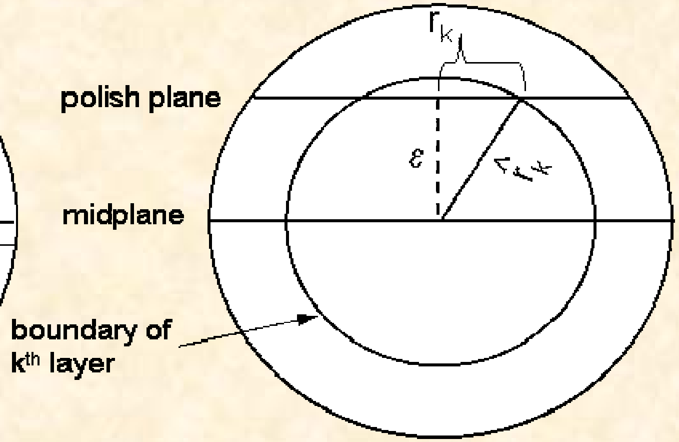


The variation in particle size causes variability in the location of the polish plane.

for a typical TRISO particle with 500 μm kernel, particle radii vary +/- ~25-40 μm.

Layer thickness measurements have systematic errors associated with off-midplane polishing that can be corrected.



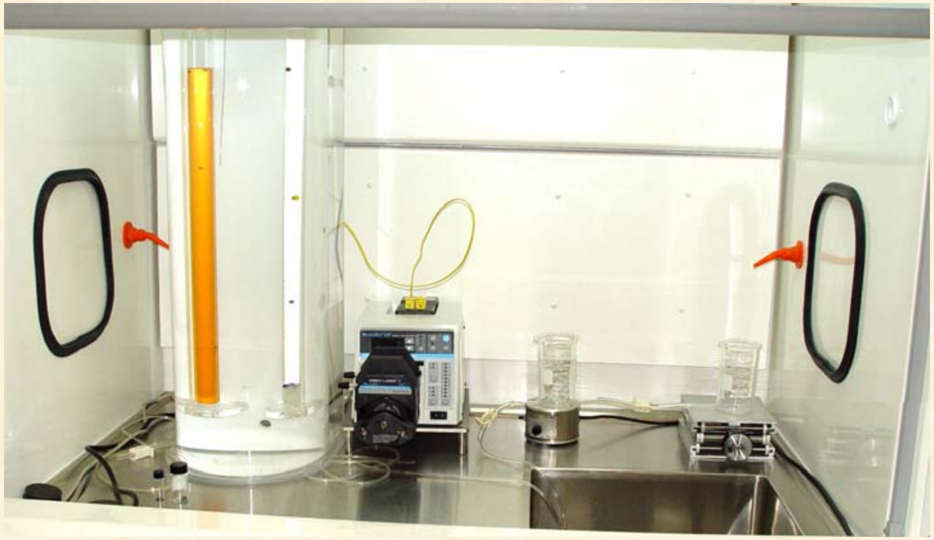


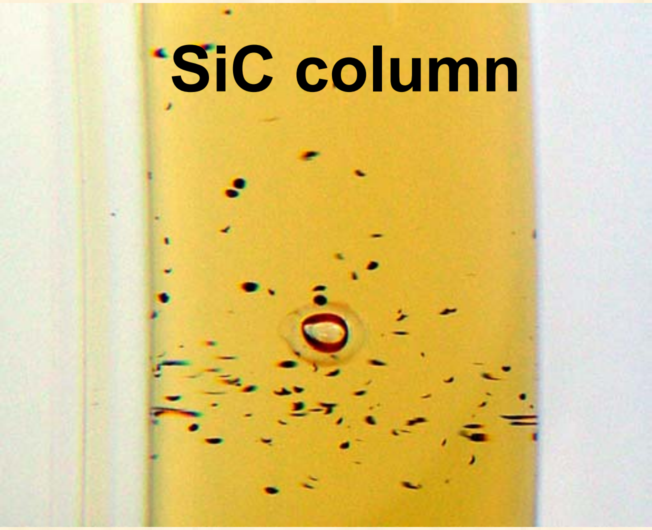
ORNL off-midplane correction:

$$\hat{r}_k = \sqrt{r_k^{\,2} + 2R_m\Delta + \Delta^2}$$

Density column

Used to determine densities of deposited layers (IPyC, SiC, OPyC)





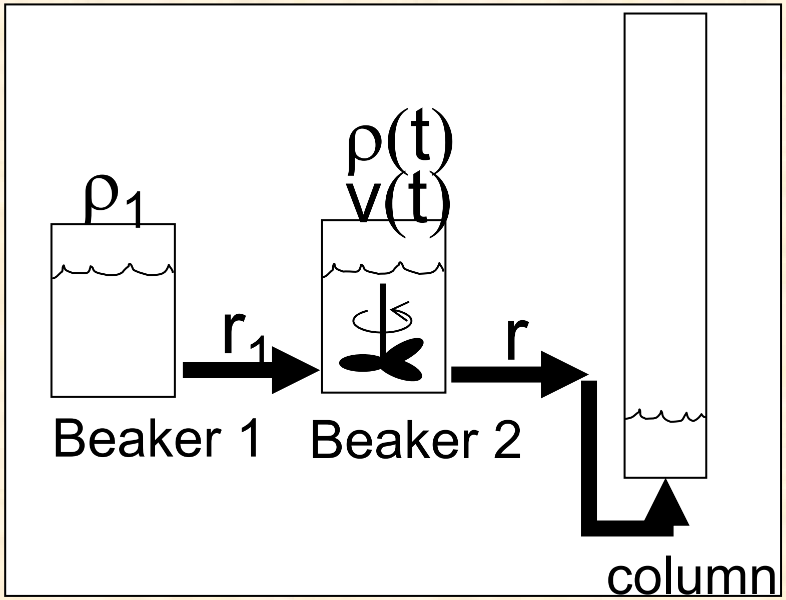
Density column created with nearly linear density gradient.

Standards of known density are placed into the column.

Drop material of unknown density into the column.

Measure position of material relative to the floats to determine the unknown density.

Construction of linear density columns



Improved column linearity can be achieved by controlling both flows.

$$\frac{d\rho}{dt} = \frac{d}{dt}\left(\frac{w}{v}\right) = \frac{1}{v}\frac{dw}{dt} - \frac{\rho}{v}\frac{dv}{dt}$$

$$v(t) = v(0) + (r_1 - r)t$$

$$\frac{dv}{dt} = r_1 - r \qquad \frac{dw}{dt} = r_1\rho_1 - r\rho$$

$$\frac{d\rho}{dt} + \frac{r_1}{v}\rho = \frac{r_1}{v}\rho_1$$

using integrating factor →

$$\rho(t) = \rho_1 + C_1 v^{r_1/(r-r_1)}$$

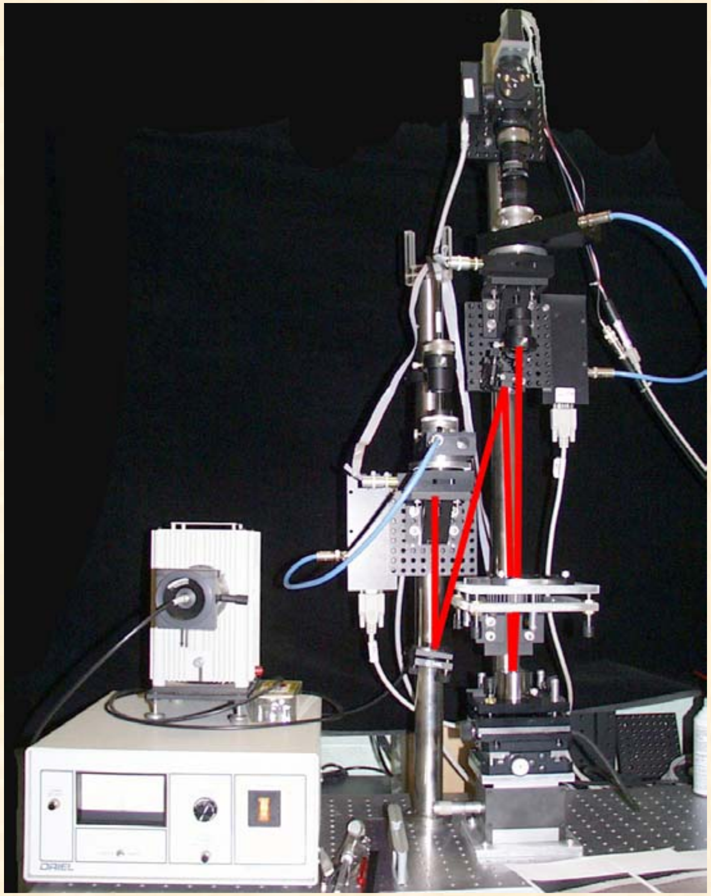
When using constant flows, a linear column can only be made if r = 2r_1.

$$\rho(t) = \rho(0) + \frac{\rho_1 - \rho(0)}{v(0)} r_1 t$$

Using this equation, linear density columns can be constructed with desired density range occurring over measurable position range.

Pyrocarbon anisotropy

A new ellipsometry microscope for measuring the crystallographic anisotropy of pyrocarbon has been developed to replace the optical polarimeter.



- Captures 8 Mueller matrix parameters at each point
- Resolution: ~4 microns
- Time/meas: 0.2-0.5 sec.
- Important parameters (accuracy ~0.002-0.005):
  - Diattenuation N
  - Retardation δ
  - Fast axis angle φ
  - Circ. diattenuation

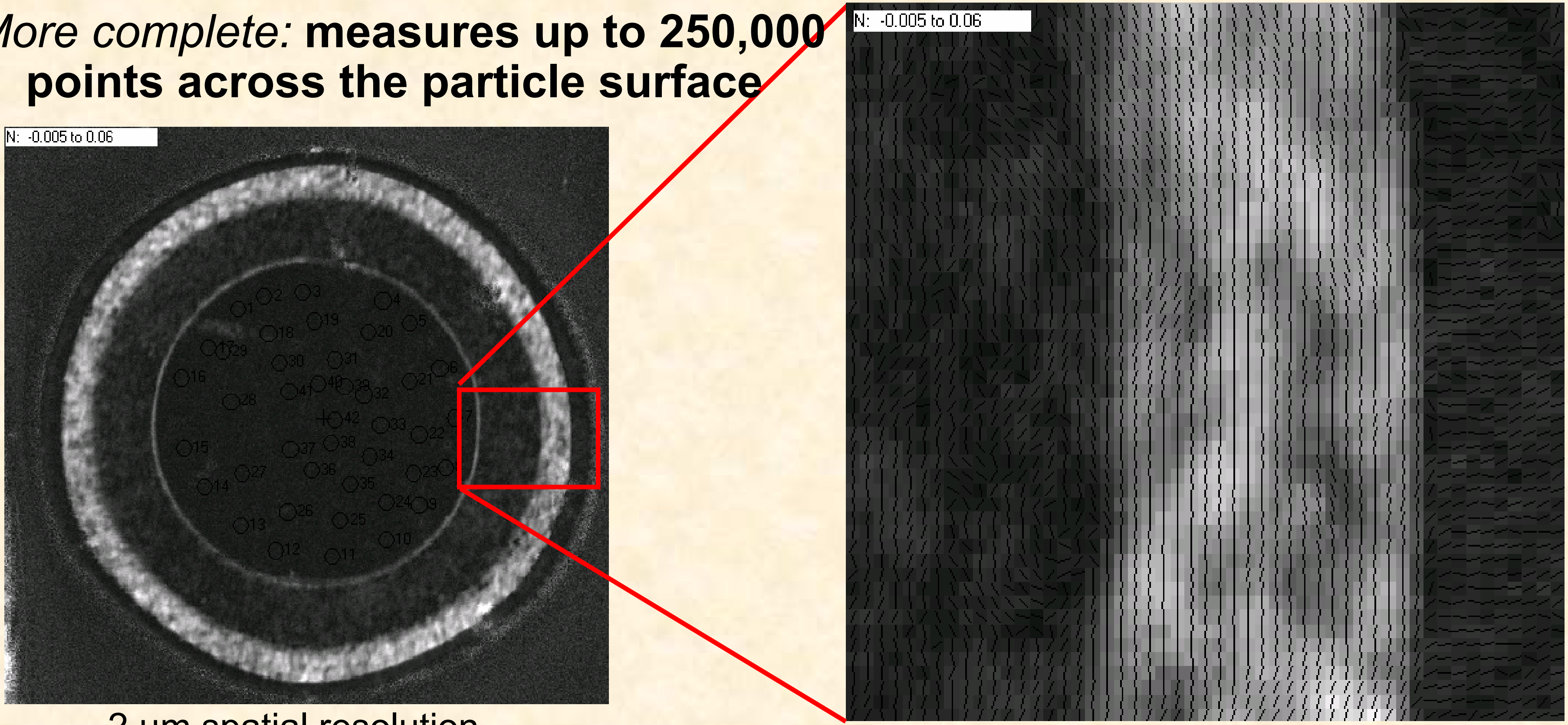
Ellipsometry microscope

More accurate: **measures the entire Mueller matrix describing polarization**

More robust: **non-specular reflection doesn't affect the accuracy**

More sensitive: **can measure anisotropies that are 10 times smaller**

More complete: **measures up to 250,000 points across the particle surface**



2 µm spatial resolution

Lines show fast axis orientation

Microwave digestion of compact impurities

Problematic impurities in compacts include:

Al, Ca, Cr, Co, Fe, Mn, Ni, Ti, V

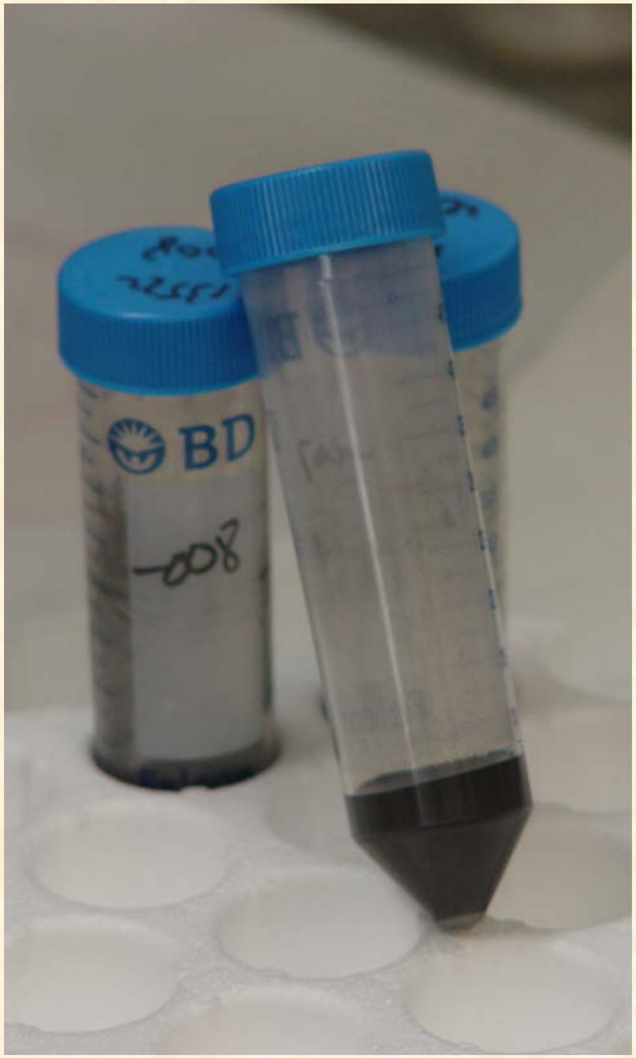
Dissolve any impurities in fluorocarbon pressure vessel in a microwave furnace (~170-210°C).

Analyze the impurity content of the leachate

(ex. ICP-AES)

Also used to determine uranium contamination and exposed uranium.

Microwave digestion



Microwave digestion of carbonized compacts with nitric acid deconsolidates the compact.

Traditional leaching takes hours, but microwave digestion dissolves the impurities in minutes.

Research on measuring impurity content has only recently been started. Preliminary results have been promising.

- In preliminary tests on doped samples, a high percentage of Fe, Mn, Ni, & Co were found in the leachate (~70-100%).

Summary

New & modified characterization methods are being developed at ORNL for kernels, coated particles, & compacts to establish a new standard for fuel characterization in the U.S.

Computer automated optical microscopy

large sample size, ease of measurement, thorough characterization

Ellipsometry microscope

improved accuracy, higher sensitivity, higher resolution

Microwave digestion

dissolution in minutes, small acid volume, ongoing evaluation of technique