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Nuclear Energy Research Initiative Annual Report

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PACIFIC NORTHWEST NATIONAL LABORATORY
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Innovative Approaches to Automating QA/QC of Fuel Particle Production Using On-Line Nondestructive Methods for Higher Reliability

Project No. 02-103

Reporting Period:

(October 2002- September 2003)

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Project Objective and Background

The successful development and deployment of Generation IV nuclear power systems strongly depends on the reliability and performance of sub-millimeter multi-layer (TRISO) particles used as fuel. The term TRISO refers to the tri-coated oxide kernel. These same types of particle fuels are used in several current and proposed advanced nuclear reactor fuel designs. The performance of the (TRISO) micro-spheres is a key component in system containment, depending particularly heavily on the properties and performance of the silicon carbide layer. Current QC schemes employ destructive testing and are not suited to in-line measurement, and are therefore economically infeasible considering future demand.

This project evaluates, develops and demonstrates Non-Destructive Evaluation (NDE) technologies that can be eventually automated to meet production speeds, provide the capability to give as close to 100% inspection as reasonably possible, and reduce the number of independent measurements needed to qualify a particle or batch. A "Quality Index" will be developed and demonstrated to give a measure of both individual particle and batch conformity. Research will consider (I) in-line measurements, (II) on-process measurements and (III) advanced off-line NDE measurements needed to qualify in-line measurements and reduce the numbers of measurements. Technologies to be evaluated include: electrical measurements (eddy-current/dielectric constant), acoustic microscopy, resonant ultrasound spectroscopy, high-resolution computed tomography, transmission and diffuse field ultrasound and optical measurements. In project year two, appropriate technologies will be down-selected for laboratory-scale testing to demonstrate their potential to satisfy throughput requirements for large-scale fuel manufacturing.

Executive Summary

This document summarizes the activities performed and progress made in FY-03. Various approaches for automating the particle fuel production QC process using on-line nondestructive methods for higher reliability were evaluated. In this first-year of a three-year project, surrogate fuel particles made available for testing included leftovers from initial coater development runs. These particles had a high defect fraction and the particle properties spanned a wide range, providing the opportunity to examine worst-case conditions before refining the inspection methods to detect more subtle coating features. Particles specifically designed to evaluate the NDE methods being investigated under this project will be specified at GA and PNNL, and then fabricated at ORNL early next reporting period. The literature was reviewed for existing inspection technology and to identify many of the fuel particle conditions thought to degrade its performance. A modeling study, of the electromagnetic techniques, showed that the in-line electromagnetic methods should provide measurable responses to missing layers, kernel diameter variability, and changes in coating layer thickness, with reasonable assumptions made for material conductivities. The modeling study for the ultrasonic methods

provided the resonant frequencies that should be measured using the resonant ultrasound technique, and the results from these calculations were published in the proceedings for two conferences.

The notion of a particle quality index to relate coating properties to fabrication process parameters was explored. Progress was made in understanding the fabrication process. GA identified key literature in this area and provided a literature review/summary. This literature has been reviewed. An approach previously applied to flexible manufacturing was adopted and the modification and development of the concepts to meet TRISO particle fuel manufacturing and QA/QC needs was initiated. This approach establishes relationships between key process parameters and part parameters, including "defects" for each manufacturing step - which in this case is a coating layer. This activity will continue in year two, when an initial evaluation will be made using available process and particle data.

Radiographic and Computed Tomography (CT) techniques were developed and refined to examine individual particles and batches of up to about 30 to 40 particles for kernel diameter, coating layer thickness and spatial uniformity. These results are essential for developing the defect library of characterized particles that will be used to calibrate the high-speed nondestructive measurement methods that are found capable of automatically detecting particles having properties outside a specified range.

The evaluated in-line inspection methods include the electrical property measurement methods traditionally referred to as eddy current and capacitance (or dielectric) in the nondestructive test methods literature. An eddy current technique was developed and evaluated on stationary particles. Good correlation was found between the eddy current measurements and the radiographically determined particle dimensions. Initial measurements on fuel compacts using the eddy current approach showed that these materials are amenable to electrical inspection and that significant coil impedance variability can be observed among different samples. A comprehensive evaluation of the capacitance measurement method was deferred into next year due to difficulties in obtaining appropriate sensors. The calculational models predict the capacitance technique to be sensitive to the kernel and SiC layer dimensions, which is somewhat lacking in the eddy current technique. Combining both the eddy current and capacitance methods is expected to compliment the individual use of each method.

The resonant ultrasound technique was evaluated using metal spheres ranging from 1.6 mm to 25 mm. This initial evaluation showed that the measurement system, as configured, could not determine the resonant frequencies for spheres smaller than about 4 mm due to a large background and relatively small sample response. Two potential solutions were identified for overcoming this limitation. The first involved exciting particles with Electromagnetic Acoustic Transducers (EMAT's) to avoid contacting the particles, which proved to be unsuccessful due to the low energy transfer from the EMAT to the particles. The second approach was to fabricate custom contact transducers matched to the mass of the particles to reduce background transducer resonances. This second approach must be deferred into the next reporting period to permit the

manufactures enough time to fabricate and test the custom transducers.

The initial evaluation for using the acoustic microscopy approach to characterize coated surrogate fuel particles has shown that only the extreme high end of the ultrasonic frequency spectrum—above 250 MHz, and perhaps as high as 1 GHz—is effective for imaging microstructural features in the TRISO fuel. The currently available ultrasonic system is not capable of operating at frequencies above 100 MHz. Therefore, it is being recommended that the ultrasonic approach to fuel particle characterization be modified for obtaining a quality signature, one that includes a combination of particle properties and not be used to image flaws as originally proposed. This evaluation will be completed early next reporting period.

On-process measurement technologies were investigated. An initial review identified five technologies that could potentially be applied: (I) transmission ultrasound, (II) Pressurized gas-coupled ultrasound, (III) Electrical impedance and capacitance measurements, (IV) Ultrasonic backscatter, scattering and diffuse fields and (V) Process Tomography (potentially using various sensing fields). These techniques were evaluated and several were found to have potential merit. Transmission ultrasound was selected for initial development during FY 04, to provide a test bed for this and other ultrasound modalities that could be used to sense in-coater "median" particle diameter, as a function of processing time.

Results from this project appear in four publications. (See the section on presentations for details).

Research Progress

Task 1. LITERATURE REVIEW

A general literature review performed as part of the proposal process that resulted in this project revealed over 30 years of work on coated particle fuels. This initial review covered the relevant topics of advanced gas reactor programs, and in particular programs reporting the fabrication and testing of TRISO particles, both nationally and internationally. The time frame included in this initial review was from about 1970 through 2001, and includes about 150 papers.

As more recent work became available throughout the year, periodic updates were made for 2002 and the first half of 2003. Over the summer months, an undergraduate student helped build a searchable electronic database for this literature, making it easier to retrieve specific papers and add new work as it becomes available. This database currently contains about 200 papers. Accumulating new papers over the remainder of this project will be a relatively small task, using the existing database.

A review article highlighting developments in the characterization and performance evaluation of TRISO fuel particles over the last four decades is being prepared and is expected to be submitted for publication in a journal sometime in 2004.

Task 2. MODELING STUDY

The objective of the modeling activities is threefold in helping to:

1. Develop the measurement methods consistent with the theory
2. Better interpret the measurements
3. Predict measurement results for a broader range of defect conditions, beyond those that can be fabricated in the time frame of this project.

In the first year both acoustic and electromagnetic models have been developed.

Resonance Computation Modeling

Resonant Ultrasound Spectroscopy (RUS) is a relatively new technique [A. Migliori and J. L. Sarrao] in which one measures the resonant frequencies of a sample of nearly ideal geometry and with known symmetry, dimensions, and mass. The resonance peaks are amplifications of waves through constructive interference within a solid at specific frequencies. A brief description of the technique is given here, however, more detailed information is available elsewhere [A. Migliori (1997), J. Maynard (1996), J. D. Maynard, (1992), R. B. Schwarz (2000), A. Migliori (1996)]. In RUS, a parallelepiped, cylinder, or sphere of the material of interest is suspended between two transducers across opposite edges (in case of the parallelepiped or cylinder). As one transducer excites the specimen by sweeping through a specified range of increasing vibrational frequencies, the opposite transducer monitors the response of the solid to the input signal and converts it to an electrical signal which is fed into a computer for analysis.

In RUS NDE techniques, defects in objects are detected on the basis of changes in the pattern of resonance frequencies relative to the spectrum of the defect-free object. In this task, we have computationally investigated implementing the XYZ algorithm of [Visscher et. al. (1991)] and the suitability of RUS as an NDE tool to detect shape defects and layer imperfections in sub-millimeter sized TRISO particles being considered as fuel for the Generation IV nuclear power plants. The representative material properties of the kernel and the layers of TRISO particles that we have used for the computational study are listed in Table 2-1.

TABLE 2-1. Representative material properties of TRISO particles

	Density (gm/cc)	Lame constant (GPa)	Lame constant (GPa)	Thickness / Diameter (m)
Z_rO_2 kernel	5.7	128.19	76.35	500
Porous carbon buffer layer	0.97	2	3.34	65
IPyC layer	1.875	5.5	10.5	35
SiC layer	3.19	77	199	35
OPyC Layer	1.825	5.5	10.5	40

Summary of Results

Fig. 2-1 illustrates the close agreement between computed results and experimental results for a 3.145 mm aluminum sphere [A. Migliori (1996)].

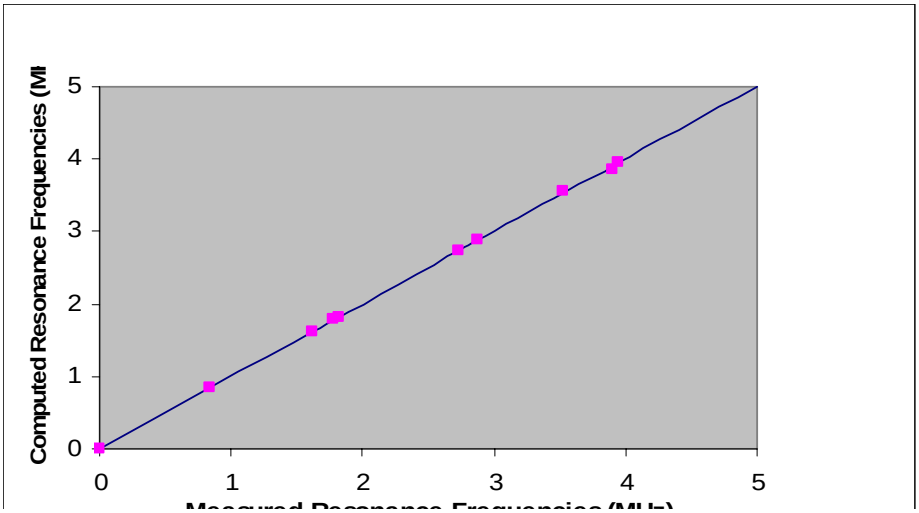

Fig. 2-1. Comparison of measured resonance frequencies with computed resonance frequencies for 3.145 mm aluminum sphere.

In order to study the effect of shape on resonance frequencies of a homogeneous sphere, we have performed calculations for different spheroids with the same volume. Fig. 2-2 shows the effect shape has on the resonance frequencies. The x-axis represents the aspect ratio between the semi-axes of a general ellipsoid. Notice the massive degeneracy of resonances in the case of perfect spheres (aspect ratio of 1). The reduction in the extent of degeneracy can be utilized as a signature of shape defect.

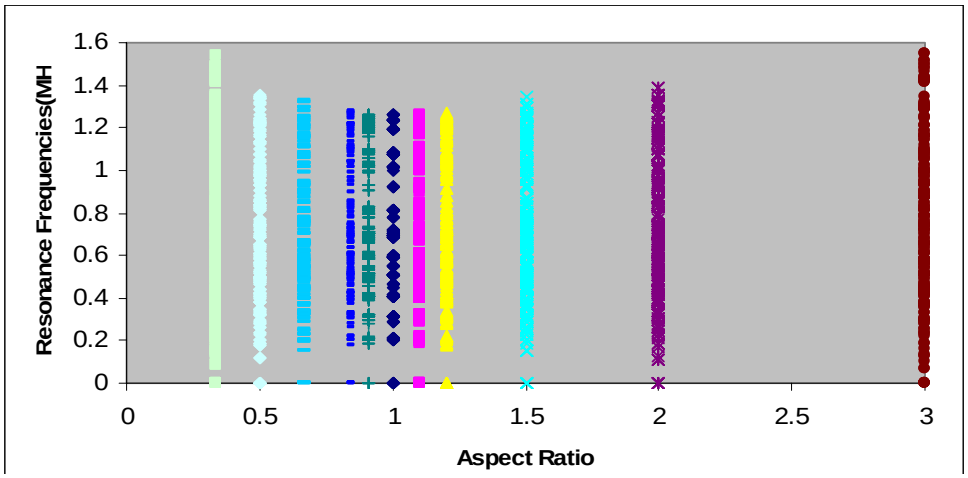

Fig. 2-2. Resonance frequencies for a family of spheroids.

Fig. 2-3 shows the variations of the first 28 free body resonance frequencies of surrogate TRISO particles caused by a 10-μm decrease in thickness of the porous carbon buffer layer. Noticeable differences in the computed frequencies can be seen when compared with those of a "flawless" TRISO particle. An important point to keep in mind is that this results in an overall diameter reduction of 20 μm. It is to be observed that this type of defect resulted in increased frequencies, relative to the frequencies of the simulated TRISO particle adhering strictly to specifications. Fig. 2-4 shows the computed frequencies for the case when the reduction of the buffer layer thickness is offset by increases in two of the layers, namely the IPyC and the OPyC layers, keeping the diameter of the TRISO particle unaltered. Interestingly, one notices that resonant frequencies of most of the displayed modes are lowered.

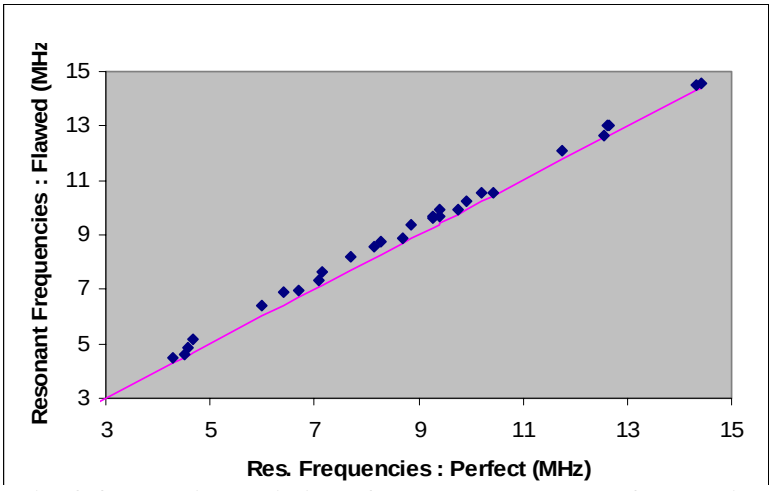

Fig. 2-3. Relative variation of computed resonance frequencies due to a 10-μm decrease in buffer layer thickness.

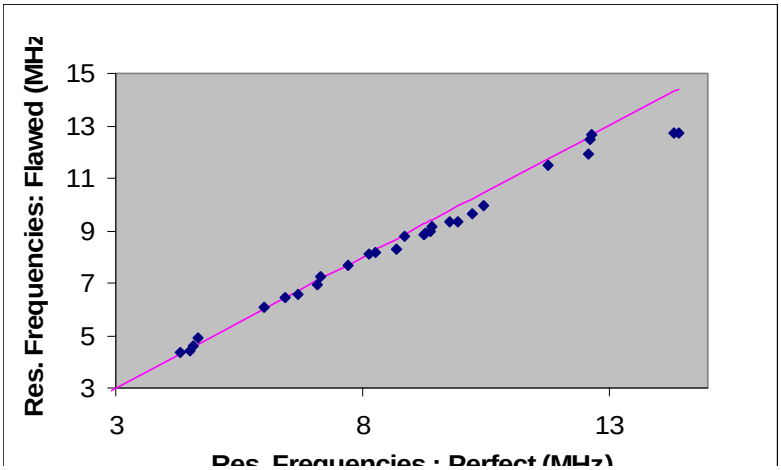

Fig. 2-4. Relative variation of computed resonance frequencies due to a10-μm decrease in buffer layer thickness along with 5-μm increase in thickness of each of the IPyC and the OPyC layers.

The results presented here do not distinguish between the different modes of vibrations. It is the surface motions of the elastic object that determine the modes of vibrations. Although we have calculated the surface displacement patterns for each vibrational mode, studies on the effects of layer "defects" on modes of vibrations have not been performed. We have computationally observed that shape defects of spheres have a strong impact on the resonance frequencies and thus RUS provides a viable alternative to look for shape defects of TRISO particles. It has also been seen that variations in layer thicknesses of TRISO particles result in noticeable changes to the normal mode frequencies. However, one must carry out further investigations to obtain a correlation between a certain layer defect and the normal mode frequency which is affected most sensitively before RUS can be used as a reliable NDE tool to detect layer imperfections in composite spheres.

Acoustic Microscopy Modeling

Since the TRISO particle layers are relatively thin, it would be very difficult to discriminate echoes from different interfaces using ultrasonic pulses when center frequencies are in the 50 MHz to 200 MHz range. In order to find alternative methods, the following modeling approach was taken, seeking defect signatures within the layers of TRISO particles.

Summary of Results

If the incident wave train is long compared to twice the thickness of the thickest layer, the various transmitted and reflected wave fronts combine in a complex way. This fact has been exploited to compute the reflection coefficient of the layered structure as a function of frequency. The numerical results plotted in Fig. 2-5 show that the reflection coefficient is sensitive to layer thicknesses. It is to be noted that when a layer thickness was decreased by an amount, another layer thickness was increased by the same amount, leaving the particle diameter unchanged.

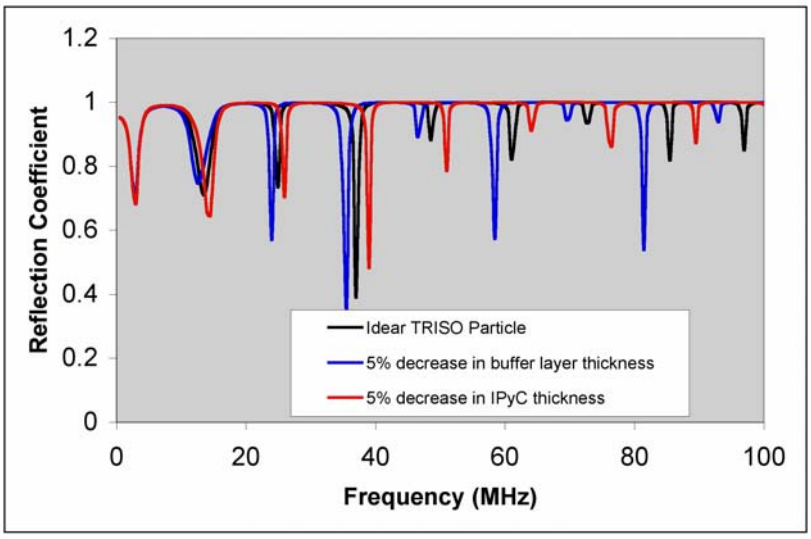

Fig. 2-5. Layer thickness variation effect on frequency dependent reflection coefficient.

Electromagnetic Modeling

Two models were developed to demonstrate how the electromagnetic properties of the TRISO fuel particle respond to an applied electromagnetic field. The first model is used in calculating the complex valued coil impedance for a changing magnetic field applied to a particle of given layer thicknesses and layer electrical conductivities. The second model focuses on calculating the capacitance change resulting from the addition of each particle coating layer, of a given dielectric constant.

Eddy Current Method

The objective of this task is to use a calculational model for predicting the eddy current response to particle defects (deviations from the specified particle) and to help define the optimal design and operational parameters of appropriate sensors. These results will become most valuable when the experimental measurements are available to verify and calibrate the models.

The software package "Electro®" by Integrated Engineering Software was purchased to calculate the dielectric constant of a TRISO particle. This software provides a solution to the 2-dimentional capacitance of a given capacitor and axisymmetric layered particle. Several different calculations were made to assist in the design of capacitor to measure particle capacitance. However, it was necessary to make several assumptions for the dielectric properties of the various particle layers, since measurement data is not available for similar coatings.

Eddy current calculations were modeled after the experimental measurements currently being conducted. The software package being used for this task is "OERSTED", also

from Integrated Engineering Software. OERSTED provides a 2-dimensional boundary element calculation of coil impedance. Coil impedance was calculated for 2-D, axisymmetric particles where only the outer pyrolytic carbon (OPyC) layer thickness was allowed to vary. Calculated coil impedance variations resulting from this OPyC variation are well above the noise level found in recent measurements and the results are shown below in Fig. 2-6.

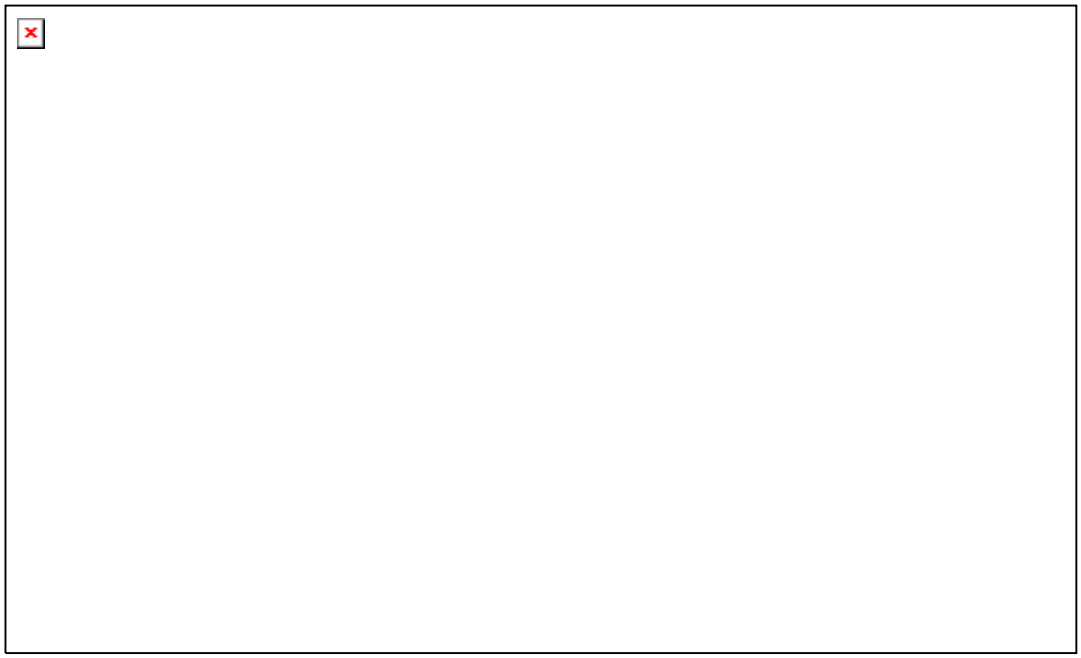

Fig. 2-6. Calculated coil impedance for TRISO coated particles of various OPyC layer thicknesses.

The largest difficulty to be overcome in obtaining relevant information from the model is the large number of unknown electrical properties for each layer, most notably electrical conductivity. Two approaches are planned to overcome this limitation. The first is to investigate particles having a single layer coating of either buffer, PyC or SiC over the kernel. This approach makes it possible to solve equations having the same number of unknown parameters as the number of measurable parameters. The second approach is to use reasonable approximations for the unknown parameters and then use several different characterized particle measurements to adjust the model to fit the measurements.

Capacitance Method

In the capacitance method a potential difference is applied between two parallel metal plates to establish an electric field. By measuring the voltage across the plates and the current flow it is possible to determine the change in capacitance and the dielectric constant due to the material, such as a TRISO particle, inserted between the plates.

Using the two-dimensional axisymmetric model, shown to the left in Fig. 2-7, the change in the capacitance due to each coating layer was computed as shown to the right of Fig. 2-

7. Notice that the SiC layer produces one of the larger changes due to its higher dielectric constant. Therefore, this type of sensor may be well suited for inspecting particles just after application of the SiC coating to determine the SiC layer quality.

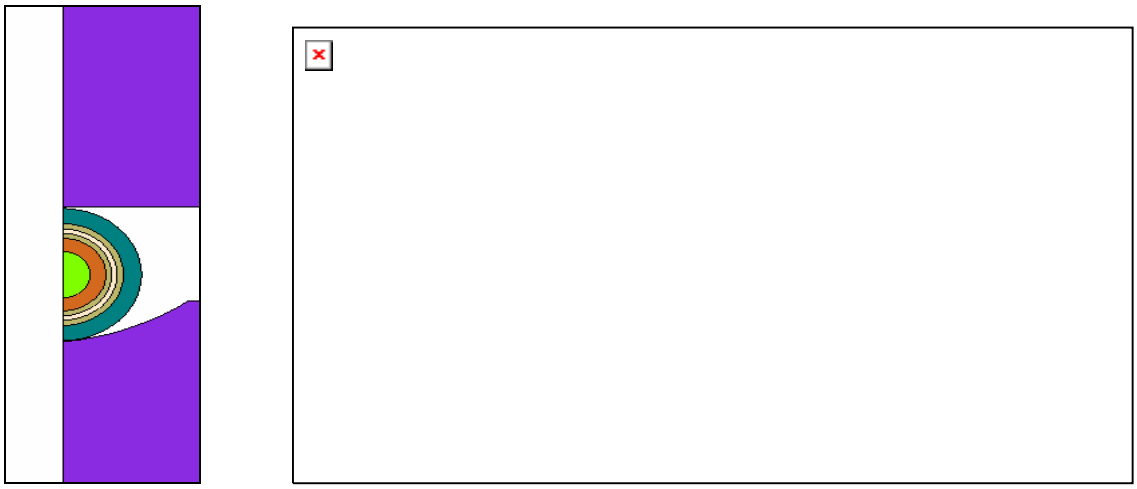

Fig. 2-7. Model of a TRISO particle inside a parallel plate capacitor (left), and the calculated capacitance change resulting from the addition of each coating layer (right).

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A. Migliori and J. L. Sarrao, "Resonant Ultrasound Spectroscopy: Applications to Physics, Materials Measurements, and Nondestructive Evaluation," (1997), John Wiley and Sons, New York.

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Task 3. DEFINE QUALITY INDEX

The quality of TRISO particles and the methods used to evaluate particles present particular challenges. Current methods are both time consuming, based on sampling, and they include destructive examination of samples. For each advanced reactor, up to 15

billion TRISO particles may be required at any one time to give a reactor core fuel-load. Refueling cycles can be as often as 18 months. It has been reported that US and German particles exhibit significant performance differences [Petti et al (2003)].

This task seeks to develop and then demonstrate a "Quality Index" that is based on non-destructive measurements. It is intended that it will be applicable to both individual particles and batches and have batch qualification/acceptance testing capability. The developing methodology seeks to relate properties to fabrication process parameters. Key fuel parameters are summarized in Table 3-1.

Process Characterization

Significant progress has been made in understanding the fabrication process. GA identified key literature in this area and Saurwein (2003a) provided a literature review/summary of much of the historic and recent literature. This literature has been reviewed. Significant papers include those by Petti et al (2003) and a special issue of a journal (Nuclear Technology, September 1977). Recent work, e.g., Sawa and Tobita (2003) considering the behavior of SiC-coated fuel particles in terms of performance, confirmed the significance of the buffer layer and identified other key parameters. Additional input from GA includes "Technical Guidance For Development of Advanced QC Methods Gas-Cooled Reactor Fuel Particles" [Saurwein (2003b)] and a "TRISO-Coating Process Summary" [Saurwein (2003c)].

Proper fluidization of the particle bed is essential for the production of high quality coatings. Key process parameters are identified as:

- Total gas flow
- Batch size - Mass/volume of particles
- Diameter of the coater (furnace)
- Bed surface area
- Design of gas distributor
- Gas concentrations
- Gas fluidization velocity
- Coating temperature
- Coating rate

Table 3-1. Summary of GT-MHR Fuel Property Specifications Important to Performance

Property	Property Type	Mean Value	Critical Region	Defect Fraction
Kernels				
C/U atomic ratio	Variable	0.50 ± 0.20	≤0.01 ≤0.20 ≤0.01 ≥0.80	NA
O/U atomic ratio	Variable	1.50 ± 0.20	NA	NA
Kernel diameter (μm)	Variable	350 ± 10	≤0.01 ≥400	NA
Kernel density (Mg/m^3)	Variable	≥ 10.5	NA	NA
Coated Particles				
Buffer thickness (μm)	Variable	100 ± 10	≤0.01 <50	NA
Missing buffer (≤15 μm)	Discrete	NA	NA	1.0×10^{-5}
IPyC thickness (μm)	Variable	35 ± 5	≤0.01 <20 ≤0.01 >50	NA
IPyC density (Mg/m^3)	Variable	1.80 – 1.95	≤0.02 <1.80	NA
IPyC anisotropy (BAF_o)	Variable	1.040	≤0.01 >1.060	NA
SiC thickness (μm)	Variable	35 ± 5	≤0.01 <25	NA
SiC density (Mg/m^3)	Variable	≥3.18	≤0.01 <3.17	NA
SiC integrity – gold spots or visible flaws	Discrete	NA	NA	1.0×10^{-3}
OPyC thickness (μm)	Variable	40 ± 10	≤0.01 <21	NA
OPyC density (Mg/m^3)	Variable	1.75 – 1.90	NA	
OPyC anisotropy	Variable	1.040	≤0.01 >1.060	NA
Missing OPyC (≤20 μm)	Discrete	NA	NA	1.0×10^{-4}
OPyC integrity	Discrete	NA	NA	1.0×10^{-2}
Compacts				
U contamination (grams exposed U/gram U)	Variable	NA	NA	1.0×10^{-5}
SiC defect fraction	Discrete	NA	NA	5.0×10^{-5}
IPyC integrity (fuel dispersion)	Discrete	NA	NA	4.0×10^{-5}
OPyC integrity	Discrete	NA	NA	0.01
Iron content outside SiC (μg)	Variable	50	≤0.01 <100	0.01
Cr, Mn, Co + Ni content (ppm)	Variable	NA	≤0.01 <240	0.01

For each coating layer the greatest influence on microstructural properties of the coating has been shown to be due to variations in:

- Temperature
- Coating gas fractions

The values for the various process parameters are summarized in Table 3-2.

Table 3-2. TRISO coating gases, coating rates, active coating gas fractions, and temperatures.

Coating	Diluent and Levitation Gas	Active Coating Gas	Mean Coating Rate (μm/min)[a]	Active Coating Gas Fraction[b] C/(C+L+D)	Nominal Coating Temperature[c]
Buffer	Ar, or Ar and He	C_2H_2	(d)	(d)	1250
IPyC	Ar	C_2H_2 and C_3H_6	≥3.0	0.25 – 0.35	1300
SiC	H_2	CH_3SiCl_3	≤0.33	0.012 – 0.021	1500
OPyC	Ar	C_2H_2 and C_3H_6	≥3.0	0.25 - 0.35	1300

Notes:

a) Mean coating thickness divided by coating deposition time.
b) C = active coating gas flow rate to coating zone (C_2H_2 + C_3H_6 for PyC coatings only), (CH_3SiCl_3 for SiC coatings only). L = levitation gas flow rate to coating zone (Ar for PyC coatings only), (H_2 for SiC coating only). D = diluent gas flow to coating zone (Ar for PyC coatings only), H_2 for SiC coating only).
c) Normal temperature in the active coating zone of particle bed.
d) Not defined.

A significant measure of coater performance and effectiveness is the "coating rate." However, the *true* coating rate is a non-linear parameter with time. In current practice what is monitored and calculated is the "*effective*" coating rate, which is defined as the mean coating thickness divided by coating time, and taken as averages for a batch of particles. This is based on sample physical analysis—post run. Methods for monitoring true median particle diameter during the process were found to be of potentially significant value and could give improved coater control.

Quality Index

A standard distribution (shown in Fig. 3-1) can be applied to a single characteristic such as a layer thickness or a particle diameter and simple metrics for quality defined. For the TRISO particles as shown in Table 1, there are numerous parameters that all contribute to the definition of an "acceptable" particle.

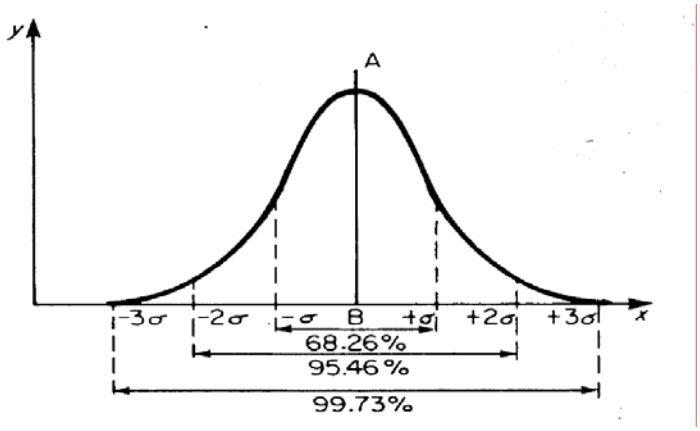

Fig. 3-1. Standard distribution.

The primary challenge for developing a metric to define "quality" for the TRISO particle is that there are multiple characteristics, each of which has acceptance bands. For a quantitative relationship to be established, correlations between the key process parameters (summarized in Table 3-2) and resulting particle parameters shown in Tables 3-1, and 3-2 must be established.

An approach is proposed for a quality index relating "process" to "part" parameters. It is developed from a methodology proposed and demonstrated for a flexible manufacturing plant by Seifoddini and Djassemi (2001). For this application, parameters were related to the performance of each specific machine and the Quality Index (QI) was proposed as a part screening mechanism.

A graphical representation for established relationships between "key process parameters" and "part parameters" for each "manufacturing step" is shown in schematic form as Fig. 3-2.

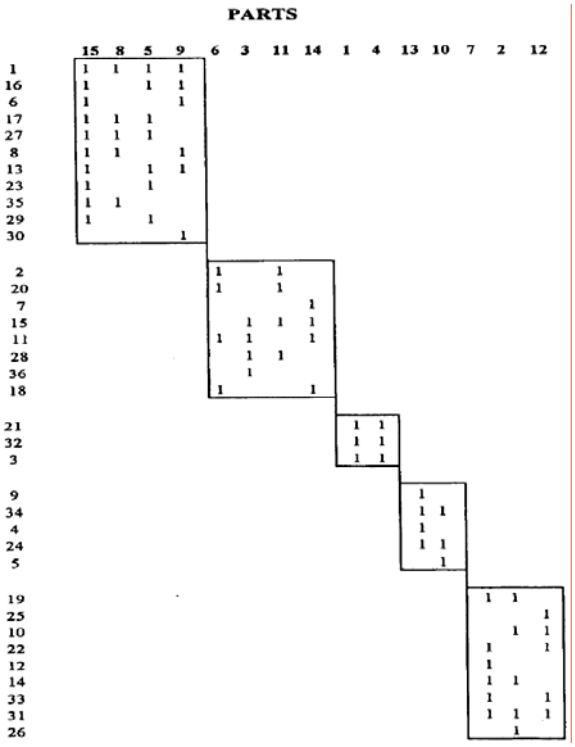

Fig. 3-2. Schematic showing groupings between "Process steps/machines"—vertical axis and "parts"—horizontal axis. Each rectangle represents a process step—1, 0 represent part acceptable, reject respectively.

The data given in Tables 3-1 and 3-2 are then used to define the rows and ranges for acceptable "boxes" for process parameters and these are correlated with directly resulting columns for particle properties, such as a layer thickness resulting from a mean coating rate, gas mixture, temperature and a duration. Each step in the process, layer, is described in terms of a "box". At the minimum for a particular coater run 1:0 are used to define "accept" - "reject" conditions measured for a particle or particles. More probably, each part-process step and desired condition is quantified in terms of deviation from nominal. For example, a thickness may be nominally 200 micron—with acceptable from 190 to 210 micron. A particle with a 200 micron layer would score "10" - for each micron off reduce the score by "1." For a key variable - layer present or missing are scored using a 10:0 scale - and this becomes a multiplier when the quality of the particle is measured.

Table 3.1 and 3.2 specify acceptable ranges for coating processes and TRISO particle layer characteristics. A series of "rectangles" are being developed that bring together these processes and particle parameters.

Each step in the process is used to develop a multi-dimensional “Box” for relationships between acceptable process parameters, and acceptable part parameters. The challenge now remains to establish, and demonstrate the relationships between measured

parameters, as determined using the NDE tools being developed under other tasks.

In addition, process parameters can potentially be directly measured using on-process measurements that are discussed under Task 8.

The work by GA has identified that, from a performance perspective, the most important defects to detect and characterize are:

- Missing buffer coating
- Heavy metal contamination
- Defective SiC
    - Spatial defects penetrating the SiC layer
    - Grain size and structure
    - Free silicon or free carbon
    - Structural flaws, such as inclusions resulting in "gold spots"
    - Impurities
- IPyC density
- IPyC thickness
- IPyC anisotropy
- OPyC anisotropy

This defect population will also be used to develop and refine the Quality Index matrix, similar to Fig. 3-2.

Additional defects that are being investigated include the lack of a common centroid for kernel and layers, and other manufacturing defects identified and included in the sample library.

Each "box" of the type shown in Fig 3.2 is then established to define acceptable and rejectable ranges. The "part parameters" are then correlated with proposed metrics obtained from non-invasive measurements – e.g. electrical parameter measures, optical measurements and calibrated against particles characterized using micro-focus radiography and applied to individual particles.

The activity under this task for FY 04 is to obtain integrated experimental data, complete the definition of the "parameter boxes" and perform an initial evaluation of the concept using individual particles, manufactured using known process parameters and characterized using micro-focus radiography.

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Seifoddini, H. and Djassemi, M. (2001) The effect of reliability considerations on the application of quality index. Computers & Industrial Engineering, 40 pp 65-77.

Task 4. OBTAIN SURROGATE PARTICLES

Dr. Lowden at ORNL shipped the surrogate fuel particles listed in Table 4-1 to PNNL in December 2002. Also within this shipment (but not appearing in the table) were several samples of uncoated ZrO_2 and HfO_2 kernels.

Table 4-1. First batch of particles from several of the early coating runs produced at ORNL.

	Kernel		
Coating Run NT-	**Material**	**Diameter (um)**	**Layers**
1	ZrO2	500	Buffer only
2	ZrO2	500	Buffer + IPyC
3	ZrO2	500	Buffer + IPyC
4	ZrO2	500	Buffer + IPyC
5	ZrO2	500	Buffer + IPyC
6	ZrO2	500	Buffer + IPyC + SiC
7	ZrO2	500	Buffer + IPyC + SiC
8	ZrO2	300	Buffer + IPyC + SiC
9	ZrO2	500	Buffer + IPyC + SiC + OPyC
10	ZrO2	500	Buffer + IPyC
11	ZrO2	500	Buffer only
12	ZrO2	500	Buffer only
13	ZrO2	500	Buffer only
14	ZrO2	500	Buffer + IPyC
15	ZrO2	500	Buffer + IPyC
16	ZrO2	200	Buffer + IPyC + SiC + OPyC
17	ZrO2	300	Buffer + IPyC + SiC + OPyC
18	ZrO2	500	Buffer only
19	ZrO2	500	Buffer only
20	ZrO2	500	Buffer + IPyC + SiC + OPyC
21	ZrO2	500	Buffer + IPyC + SiC + OPyC
22	ZrO2	500	Buffer + IPyC + SiC
23	HfO2	500	Buffer + IPyC + SiC + OPyC
24	ZrO2	500	Buffer + IPyC + SiC + OPyC

The research teams at PNNL and ISU used the above surrogate fuel particles to investigate and develop the NDE methods in FY-03. These particles were uncharacterized and found to have a "high defect fraction" as they were produced during the early surrogate coating development phase at ORNL. These facts simplified the NDE measurement development task by making defective particles more abundant and contain a wide variety of different defect types. On the other hand, this made finding the *ideal* particle, one that could be used as a standard for measuring everything else, difficult.

ORNL provided additional surrogate fuel particles late in the fourth quarter. As summarized in Table 4-2 (and defined in greater detail in the appendices), the shipment included 11 different coating runs, including particles with buffer only, buffer+IPyC, and buffer+IpyC+SiC coated on ZrO2 and HfO2 kernels. Specific dimensions (like coating thickness) and other properties of these particles were basically unknown, and they were categorized as "available leftovers" from other work. Optical measurements on a small sampling have shown diameter variations in excess of ±5 percent.

Table 4-2. Second batch of particles from later surrogate coating runs produced at ORNL.

Coating Run	Kernel			Charge In (g)	Layers
	Cone ID (mm)	Material	Diameter (um)		
NT-52	25	ZrO2	500	10.008	Buffer + IPyC + SiC
NT-64	25	ZrO2	500	10.009	Buffer + IPyC + SiC
NT-74	50	ZrO2	500	54.5	Buffer + IPyC + SiC
NT-75	50	ZrO2	500	54.5	Buffer + IPyC
AGR-08282003-1	50	ZrO2	500	54.5	Buffer
AGR-08282003-2	50	ZrO2	500	54.5	Buffer
AGR-08282003-3	50	ZrO2	500	54.5	Buffer
AGR-08292003-1	50	ZrO2	500	54.5	Buffer
AGR-09022003-1	50	ZrO2	500	54.5	Buffer
AGR-09032003-1	50	ZrO2	500	54.5	Buffer + IPyC
AGR-09042003-1	50	ZrO2	500	54.5	Buffer + IPyC
HfO2, Buffer only	50	HfO2	500	92.7	Buffer
HfO2, B + IPyC	50	HfO2	500	92.7	Buffer + IPyC

A simplified approach to analyzing *defects* in surrogate TRISO particles is also underway. In this approach, instead of working with the fully coated TRISO, only the kernel with a single layer is investigated. This reduces the number of parameters being investigated simultaneously, for each measurement technique, at any one time. Particles are being designed (beginning next quarter) to have only a single coating layer of either porous carbon buffer, SiC or PyC. This will focus the measurements on only the simultaneous parameters for a single-layer coating, including thickness and density. Investigating the full TRISO is necessary but also allows competing effects to confuse or obscure one another. Once the NDE measurement sensitivity to the properties of a single-layer is understood then the combined effects from each layer in the full TRISO will become easier to deconvolve.

Task 4a. PARTICLE CHARACTERIZATION

Initially, the plan for characterizing particle density distribution was to use 3-D CT (computed tomography) to sort each particle. After processing the large volumes of high-resolution CT data (~1 GB) for each particle and considering the 90 minutes it takes to mount, scan and reconstruct an image for a single particle, it quickly became apparent that a faster and more efficient method was necessary to obtain the full spectrum of defect types necessary for a QA/AC project involving over one hundred different types of defects. Even though CT techniques can also provide detailed 3-D information for more than one particle, as shown below in Fig. 4a-1, this much information is not necessary in the initial screening phase, where many particles from several different coating runs must be examined for defects in their coating layers. The CT analysis is more suitable to gaining a comprehensive, multidimensional view of a particular particle once an estimate of the type and magnitude of its defects are established.

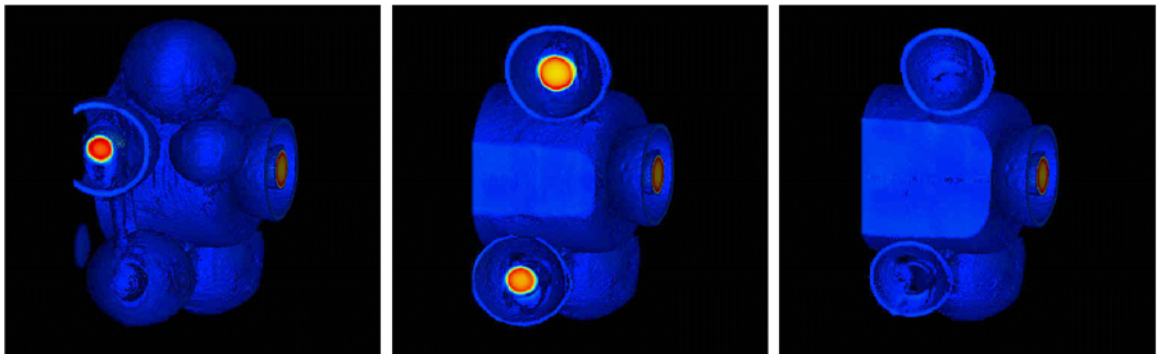

FIG. 4a-1. Three different slices through a 3D image of a CT scan of several particles attached to the tip of a glass pipette.

Digital radiography was found to be a faster and more efficient approach to rapidly assessing a batch of particles and selecting only those with the most representative features of defects thought to degrade particle fuel performance in a reactor. The radiographic presorting approach can scan 20-30 particles in only a few minutes and produce images similar to the photo of Fig. 4a-2 below. Also, by presorting with digital radiography at high geometric magnification, typically 30X, images with a 3-micron pixel size are possible using the system developed for this purpose at ISU's Center for NDE. As the radiograph of Fig. 4a-2 shows, many particles are *stacked* in a thin-wall tube, making it possible to track each particle's identity relative to the resulting digital radiograph.

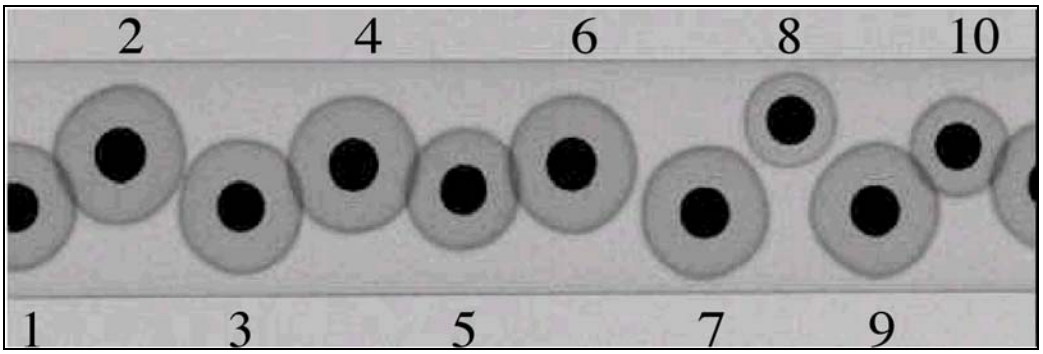

FIG. 4a-2. Digital radiograph of ten particles in a horizontal glass pipette (of 10-micron wall-thickness) illustrates the use of digital radiography as an initial sorting tool for finding anomalous particles. Notice particles 5, 8 and 10 are relatively small and particle 9 has an irregular shape.

Several particles with different coating layer defect types were found using the digital radiographic presorting approach, and now that the coating capability support at ORNL is becoming routine, faster characterization is becoming essential to keeping pace with the increase in production. The following figures contain specific examples of a few selected coating defect types discovered in using the radiographic presorting approach developed under this project to reduce characterization time.

The particle images in Fig. 4a-3 were flagged as anomalous by the radiographic sorting technique for their dissimilar sizes. A digital image analysis of this figure shows that the OPyC layers of the two particles are about the same, whereas the particle to the left has a thinner buffer and SiC coating. Furthermore, comparing this image with others reveals that the SiC coating on the particle to the right is thicker than *normal*.

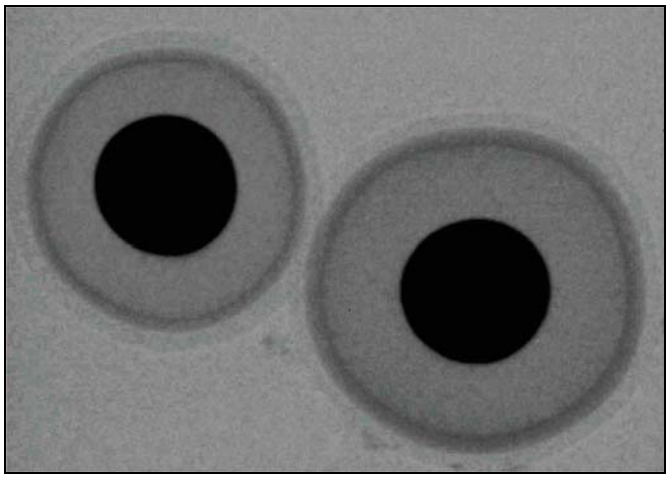

FIG. 4a-3. The left particle in this image shows a thick OPyC, thin buffer and SiC, while the particle to the right shows a relatively thick SiC.

Another important observation from radiographic presorting was that average diameter is not always a valid approach to finding missing (or thin) layers. Fig. 4a-4 is an example of a defect where the overall diameter of two particles is normal, however, their OPyC and SiC layers are thin or missing. Therefore, each layer must be taken into consideration, not just the outer diameter as is done in a sieving process.

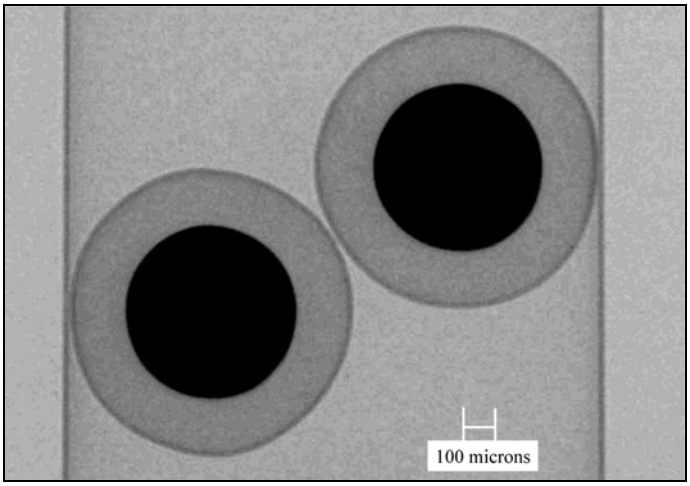

FIG. 4a-4. Both particles are about the correct size, 900-micron. However, the outer carbon layer is missing and the SiC layer is thin. These represent defects that would go undetected by a sieving operation.

Another common feature observed by radiographic imaging was non-uniform coating thickness, which typically results in a nonspherical shaped particle. In many cases the buffer coating is applied non-uniformly to a small fraction of the particles, which resulted in images similar to the one shown in Fig. 4a-5.

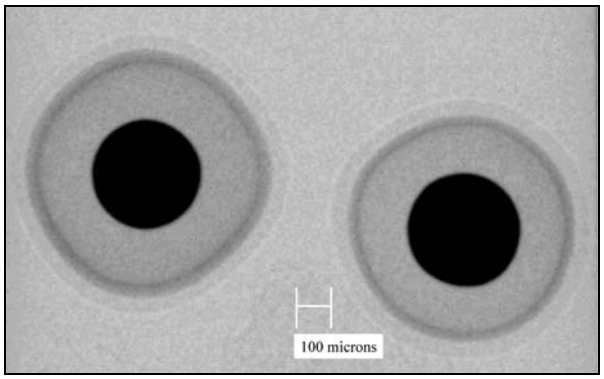

FIG. 4a-5. Irregular shaped particles, which may result in unsafe stress and strain relationships among the TRISO layers under reactor conditions.

This same micro-focus radiographic technique as described here will be continually used throughout the project in building the flaw library of Task 9.

Task 5. INTRODUCE DEFECTS IN SURROGATE PARTICLES

Although initially scheduled to begin in July 2003, this task has been deferred until particles can be coated with flaws better representing those most likely to occur in fabricating TRISO fuel. Surrogate particles now being defined for coating will contain many of the defects found in particles known to fail under test reactor conditions. Examples include a thin or missing coating layer, and variations in the density and microstructure of the PyC layers.

This task will, however, be evoked as necessary further into the project to make particles available with defects that cannot be found (or fabricated) having the necessary defects required to develop a standard flaw library containing multiple particles for each defect type known to affect fuel performance. Examples of how this may be implemented are to use compression to induce cracking in the SiC layer or perhaps using a pulsed laser to "drill" micro-pores in the SiC layer.

Task 6. DESIGN, DEVELOP STATIONARY PARTICLE NDE METHODS

The purpose of this task is to investigate four different NDE methods and develop the most promising techniques for demonstrating in-line QA/QC for each coating process of the TRISO fuel coating sequence. The four methods include:

- EM method
- UT Resonance method
- Acoustic Microscopy
- Optical Method

Each of these methods uses a different physical principal to interrogate characteristic material properties within the TRISO particle.

The optical and EM methods use electromagnetic energy to interrogate coating properties and are capable of functioning at the high throughput speeds (approaching 200 particles per second) required for 100 percent inspection. The optical method uses light reflected from the surface to provide diameter, shape and surface breaking features like cracks and pits. The EM method includes techniques using electromagnetic induction and capacitance to interrogate coating properties below the surface, including coating thickness and microstructural features associated with unique electrical conductivity signatures.

The ultrasonic methods (UT Resonance and Acoustic Microscopy) were chosen to interrogate the microstructure of each coating layer. These methods operate at inspection

speeds slower than the optical and EM methods and therefore could possibly be used in a batch sampling inspection mode (less than 100 percent inspection) to provide a statistical measure of total defect fraction.

The following is a progress summary for each of these NDE methods.

EM methods

Progress in the first half of this reporting period focused on designing, developing and characterizing the sensors thought to provide the greatest sensitivity to the various defect conditions known to occur in particle fuel. Essential to the performance of each design is the test procedure used in the evaluation. One of the foremost challenges in making each design functional was developing a procedure to insert and eject sub-millimeter sized particles without disturbing the sensor, making it possible to accurately demonstrate measurement repeatability. Approximately 10 different sensor designs (including both eddy current and capacitive sensors) were evaluated and refined before arriving at a technique that provides the level of accuracy necessary to reliably detect some of the relevant differences among the available particles.

One of the earliest eddy current sensor designs showed that this technique is clearly sensitive to particle diameter. However, at this early phase in the project the available particles were basically uncharacterized, with the outside diameter being the only known property. An example impedance plane plot, showing the eddy current coil response, is shown in Fig 6-1. The particle labels in the legend of Fig. 6-1 distinguishes between large and small diameter particles by the beginning letter of "L" or "S", respectively.

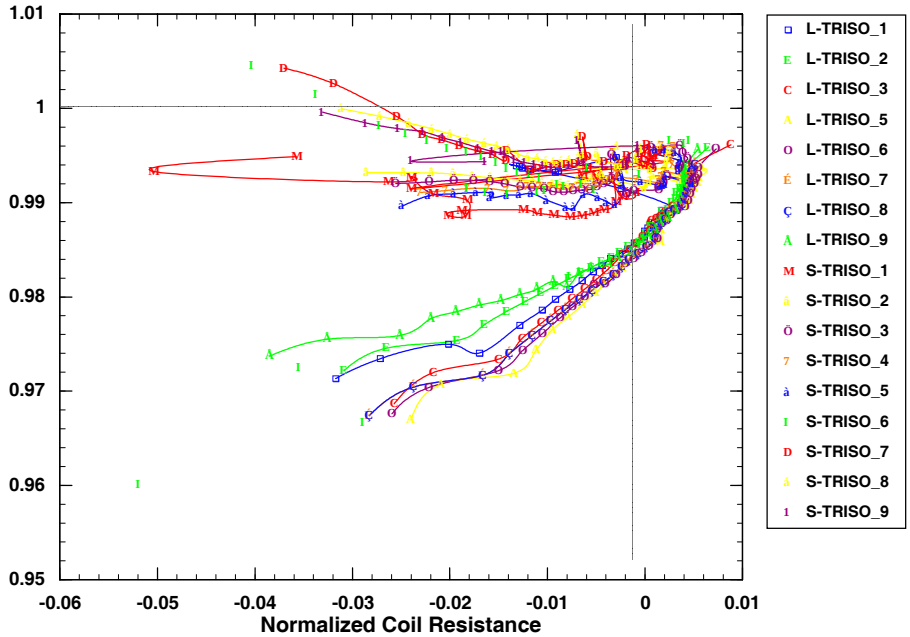

Fig. 6-1. Normalized coil impedance of 18 different TRISO surrogate fuel particles, separately placed in the test coil.

As the development of X-ray analysis techniques advanced throughout the first half of this reporting period, the inspection analysis time to determine particle density distribution diminished significantly, primarily in the second half of this reporting period. The radiographic results provided sufficient information to begin relating the eddy current measurements to internal particle properties.

Fig. 6-2 shows how the dimensional results derived from radiography relate to the coil resistance measurements. Neglecting one or two outliers, a linear relationship can be seen between either the particle sizes or the combined buffer/IPyC coating thickness and the eddy current coil resistance, as the particles were placed at the center of the coil.

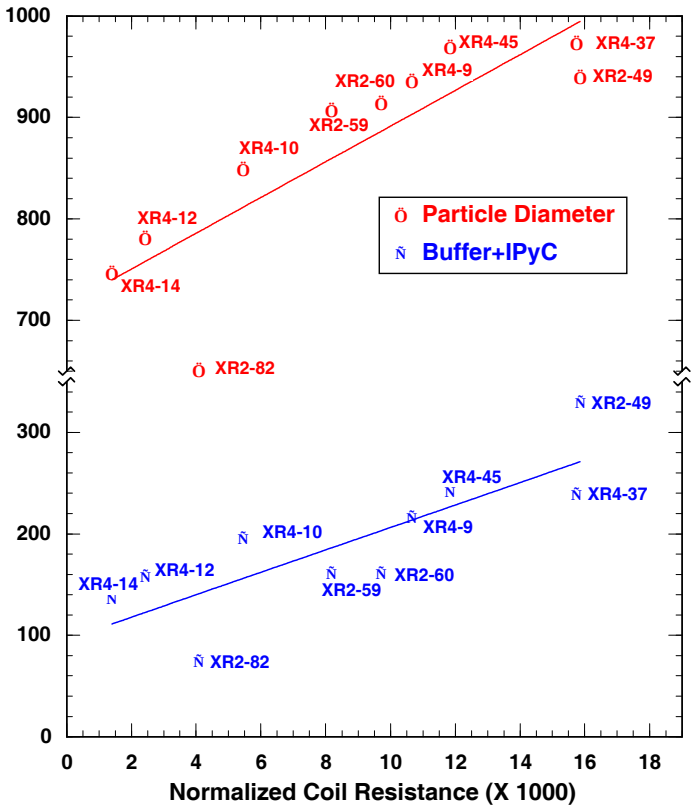

Fig. 6-2. Normalized coil resistance plotted against particle diameter and the combined buffer plus IPyC layer in fully coated ZrO_2 surrogate TRISO particles.

The next step is to calibrate the eddy current method and establish eddy current signatures for each flaw type expected to affect particle fuel performance. The effect each individual layer, as well as a combination of various layer properties, may have on the coil impedance will be fully investigated in the first quarter of FY-04. This investigation will include missing (or thin) layers and layers of different densities. The first activity in conducting this investigation will be to specify the coating parameters for

a matrix of coating runs required to produce the necessary particles. This activity is discussed further under Task 9, developing the standard flaw library.

The eddy current technique was also used to assess the electrical properties of *fuel compacts*–elements containing TRISO fuel particles inserted into a reactor in the form of fuel bundles. Fuel compacts are constructed from thousands of TRISO particles bound together in a carbonaceous matrix. Three different fuel compacts were available for this evaluation. The compacts came in three different lengths, two were several times longer than the sensor coil and one was approximately twice the coil length. Each fuel compact was placed inside a coiled wire having an inside diameter slightly greater than the compact, as shown in the photograph of Fig. 6-3. The coil was excited over a broad range of frequencies as denoted in Figs. 6-4 and 6-5.

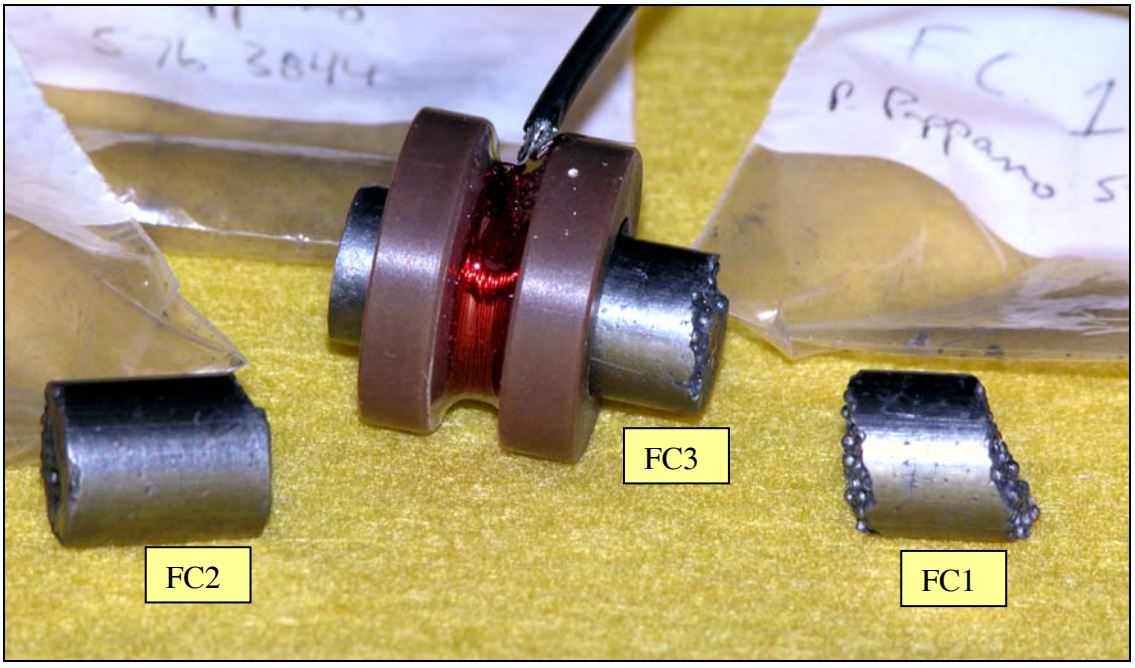

Fig. 6-3. Fuel compacts assessed using an eddy current coil. Moving the coil along the length of a full-size compact allows the coil to interrogate the electrical properties as a function of position.

The coil impedance (shown in Figs. 6-4 and 6-5) for each compact is distinctly different. In the case of fuel compact sample FC1, the reduced coil impedance could be partially due to its smaller length, because it was a broken piece, about 1 cm in length, almost twice that of the coiled wire—allowing end-effects to influence the result. However, the length of samples FC2 and FC3 should not have affected the coil impedance measurement because they were both several times longer than the coil.

The impedance of a coil surrounding a fuel compact is expected to depend on many different material properties of the compact. The most dominate properties include: compaction density, chemical composition of the matrix material, TRISO particle density

(number of particles per unit volume), void-fraction, cracking, and depending upon the conductivity ratio between the matrix material and the TRISO particles, the average TRISO particle conductivity. Without additional testing, using radiography and perhaps destructive metallographic techniques, it will not be possible to determine the principal cause(s) of these observed differences.

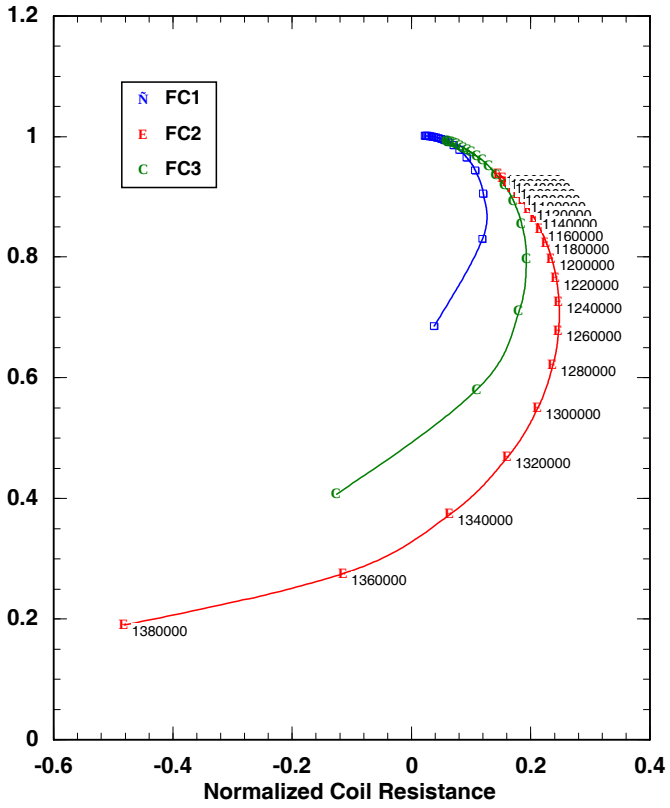

Fig. 6-4. Coil impedance normalized to the empty coil as a function of frequency ranging from 1 to 1.38 MHz.

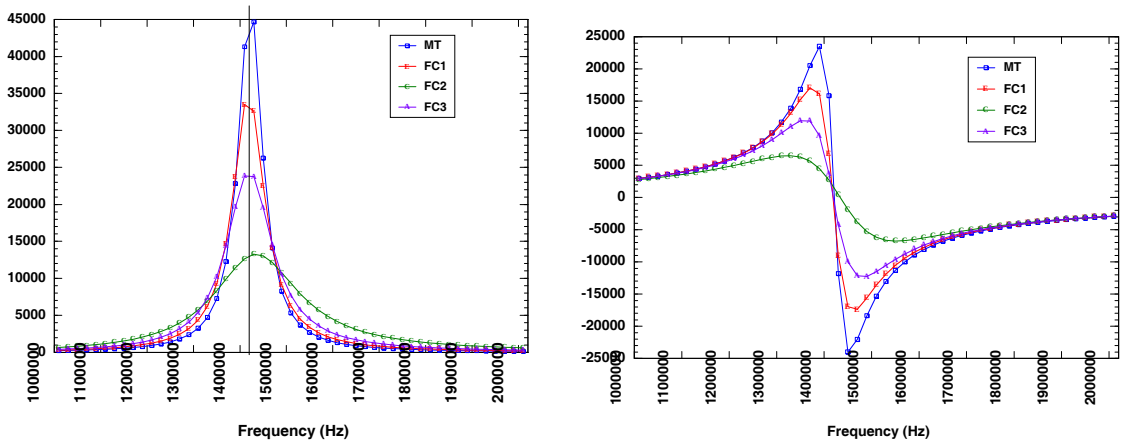

Fig. 6-5. Coil resistance (left) and reactance (right) as a function of frequency for three different surrogate fuel compacts.

In addition to the eddy current techniques mentioned above, another EM method, a capacitive measurement technique, is also being evaluated to determine the extent to which it can provide the dielectric constant of a TRISO particle that is coupled through the electric field. The development of this technique progressed slower than the eddy current technique because of the experimental difficulties involved in measuring such a small impedance change associated with mm sized particle properties, relative to a large background signal associated with the test assembly. A new design, requiring the purchase of a custom sensor, is expected to circumvent this troublesome measurement problem and will be evaluated early in next reporting period.

UT Resonance methods

The goal of this subtask is to determine the feasibility of utilizing resonance measurements to determine the physical properties of TRISO particles. The characteristic resonances of rigid bodies contain information regarding their structural properties and shape. As part of these properties the structure and make up of layers affects the characteristic resonance frequencies. In addition, the elastic moduli contribute to the resonant frequencies of the body. In principal, all of these parameters can be determined. However, if several parameters change simultaneously or affect the resonant frequency in the same way, then determining the specific property that changed can be problematic. For QA/QC, the resonance technique may be ideal, due to its high sensitivity to several important properties of the TRISO particles such as thickness of layers and composition of each layer. The results will be utilized to help determine the quality of TRISO particles during production.

This task has three activities:

1. A literature review to assess current resonance theories for predicting the resonant frequencies of spherical particles.
2. Calculations of the resonant frequencies of TRISO particles to predict the expected changes in frequency for various changes to the particle layers.
3. Experimental measurement of the resonant ultrasound spectra of spherical particles to determine the limits of the current system and to provide guidance in developing alternative measurement methods if needed.

Activity 1 and 2:

The literature review was completed under Task 1 and the modeling work is described as part of the modeling study of Task 2.

Activity 3, Experimental Measurements:

In FY 03 studies were performed to determine the sensitivity and frequency limits of the current ultrasonic resonance system at PNNL using the standard configuration. Steel ball bearings were used with diameters of 1/16”, 1/8”, 1/4”, 3/8” and 1”. The resonances for the 1/16” (1.5 mm) sphere were barley visible due to background from the transducers and the electronic system. The intermediate conclusion from this initial look shows that

the system, as configured, will have difficulty obtaining resonant spectra for the smaller TRISO particles due to a large background and the relatively small size of the particles.

To overcome this limitation of the commercial system we have pursued two paths:

1. Alternative means of transduction to eliminate contact effects utilizing electromagnetic acoustic transducers (EMATs).
2. Custom fabrication of contact transducers to eliminate or reduce background due to transducer resonances.

Path 1 EMAT Transduction:

Several specimens were sent to Dr. Ward Johnson of the National Institute of Standards and Technology (NIST) in Boulder, CO for analysis. This approach was to eliminate the cost associated with establishing this EMAT Resonance measurement capability at PNNL. These transducers rely on the interaction between the eddy current on the surface of the materials and a static magnetic field. Results showed that the electronic resonances of the system overshadowed the potential resonances of the surrogate TRISO particles. The question still remains that even if the conductivity is high enough will one be able to adequately take advantage of this transduction mechanism?

Path 2 Custom contact transducers:

Contact has been with researchers at LANL (Albert Migliori) and at the University of Tennessee at Knoxville (Veerle Keppens) to determine how to push the limits of the commercial system that we currently utilize for resonance measurements. Both scientists indicate that specific transducer configurations can be constructed that will minimize the interference from transducer resonances. Custom designs from LANL and the University of Tennessee have been considered as well as commercially available transducers from Dynamic Resonance Systems.

Future development work on this method of inspection has been deferred until the acquisition or manufacture of custom transducers and measurement on surrogate TRISO particles has demonstrated adequate extension of the capability of the commercial device owned by PNNL.

Acoustic Microscopy

This task was designated to evaluate an acoustic microscopy approach to characterizing coated surrogate fuel particles—determining the degree of penetration and feasibility of later acquiring measurements such as layer thickness, material properties, and detecting localized changes in microstructure. Initially, before coated particles were available glass spheres were examined using this method. Working at 50-MHz it was possible to see gross feature differences between a damaged and an undamaged glass sphere. The diameter of the sphere was also determined to within about 50 μm.

As surrogate fuel particles became available they were ultrasonically imaged both at PNNL and at Matec Micro Electronics. At PNNL a 100-MHz transducer was used and Matec staff used a transducer operated at 105-MHz. The PNNL system had a 30-μm lateral resolution and the Matec resolution was unspecified. Matec pursed the PNNL suggestion of using 60 °C water to couple the transducer to the particle. The transducer used by PNNL was a Panametrics V3346, 100 MHz, 3-mm diameter aperture, 6-mm focal length in water. The measured peak frequency from the samples was at 98 MHz. The transducer used by Matec was described as having a peak frequency of approximately 105 MHz and a 4.3-mm focal length in water. C-scan images obtained by PNNL and Matec had step sizes of 12 and 5 mm, respectively. These experiments demonstrated good penetration into the buffer layer, however, it was also determined that more work would be necessary to image features of interest.

As this work progressed, preliminary measurements confirmed the need to work at the extreme high end of the frequency range for ultrasonics—above 250 MHz, and perhaps as high as 1 GHz. It may be possible to work in this frequency range, but such an effort requires a larger investment than was proposed for this task. The reason it is necessary to work in this higher frequency range is to have wavelengths short enough to image and resolve the tens-of-micron size defects important to the reliability of TRISO coated particles.

However, by removing the defect imaging requirement and just detecting conditions indicative of a material properties deviation from those of a standard, it may be possible to obtain valuable results. This position is based on calculations suggesting that measurements using currently available techniques, at 50 MHz, may provide a solution and early in the next reporting period it will be evaluated.

Optical Methods

The task objective is to first review the commercially available systems for high-speed optical surface characterization systems for both batch and single-stream particle flaw inspection. A secondary objective is to explore the characterization capabilities that optical image analysis may offer if state-of-the-art technology were combined with emerging technology to potentially fill any existing gaps in realizing the imminent high-speed fuel particle inspection requirements.

An extensive review looking for existing commercially available optical inspection systems was completed. This review established that commercial, high-speed particle size and shape characterization systems are inadequate for TRISO fuel particle inspection. Dr. John Hunn (at ORNL) came to the same conclusion in his quest for an existing high-speed particle counting and sizing system. Dr. Hunn is now developing a system to meet his immediate requirements regarding sizing, based on off-the-shelf components.

To provide for the more advanced needs of the future, where high-speed coating surface flaw detection and characterization capability will become essential, an investigation into

a method that uses automated digital image analysis was initiated at PNNL. This type of inspection is expected to automatically detect surface features such as micro-pores and fissures that have been found to develop during fabrication and reported to degrade particle fuel performance. Activities this reporting period was limited to planning and system design. At the conclusion of this task, next reporting period, the capability of this inspection method will be established along with estimates for further development and demonstration.

Task 8. ON-LINE PROCESS MEASUREMENTS

This task has been considered as two elements: (a) a review and assessment of potential on-line measurement technologies and (b) preliminary results obtained with transmission ultrasound on a model coater.

(a) Review and assessment of potential on-line measurement technologies

An initial review of technologies for potential application to give advanced coater monitoring was performed. The assessment started from two published reviews by Workman et al (1999), which looked at process chemometrics, and one by Tayebi et al (2001), which considered measurement techniques for validating CFD models for multi-phase reactors. In addition various books on the emerging topic of process tomography were reviewed [e.g. Williams and Beck (1995), Plaskowski et al (1995)]. The techniques identified were then investigated further using the data base "Web of Science."

Five technologies that could potentially be implemented to give on-process monitoring for a coater-system of the type used for TRISO particle fabrication were identified:

- Transmission ultrasound
- Pressurized gas-coupled ultrasound
- Electrical Impedance and capacitance measurements
- Ultrasound backscatter, scattering and diffuse field
- Process Tomography (using various sensing fields)

Transmission Ultrasound

Transmission ultrasound has been successfully demonstrated on an autoclave, fluidized beds and other reactors by Soong (1995-2000) [e.g. Soong et al (1997), Soong et al (1999) and other groups [Bond (1998)]. This work has demonstrated that high temperature operation is feasible and radiation resistant materials for transducer fabrication have demonstrated performance in various nuclear applications. For example acoustic emission sensors, based on piezoelectric elements, have been deployed on the primary pressure vessels of nuclear submarine power reactors.

A schematic for a system on an autoclave reactor is shown in Fig. 8-1

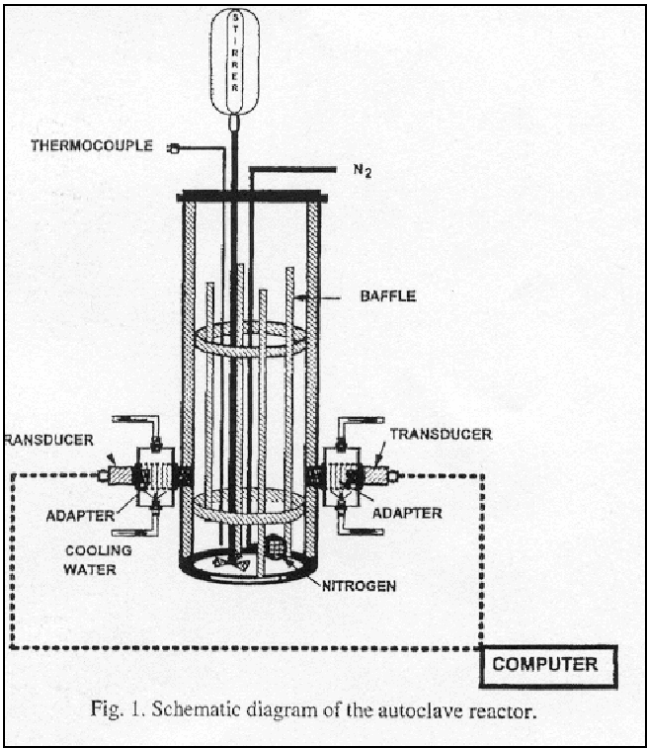

Fig. 8-1. Example of an autoclave reactor with transmission ultrasound [from Soong et al (1997)].

In addition there is significant literature that reports attenuation of sound in gas-solid suspensions [Gregor and Rumpf (1976)], velocity of sound in fluidized beds [Gregor and Rumpf (1974), Roy et al (1990)] and various implementations including in-line particle size and granular material characterization measurement in flowing bulk solids [Weir (2001), Tallon et al (2003)].

Pressurized gas-coupled ultrasound

The use of high-pressure inert gas has been demonstrated for a limited number of demanding non-contact inspection systems. The use of gas removes the requirement for a liquid couplant and through operation at elevated pressure this gives a factor of 5 improvement of resolution, when compared with the same frequency operating in water. Reflection, gas-coulpled acoustic-microscopy was demonstrated by Wickramasinghe and Petts (1980) and a transmission microscope was developed by Bond (1992). More recently, gas coupled ultrasound has been implemented for the characterization of compaction responses for porous membranes [Reinsch et al (2000)].

An example of a gas-coupled microscope image of a coin and the variation of attenuation as a function of pressure for nitrogen as shown as Figs 8.2 and 8.3, respectively.

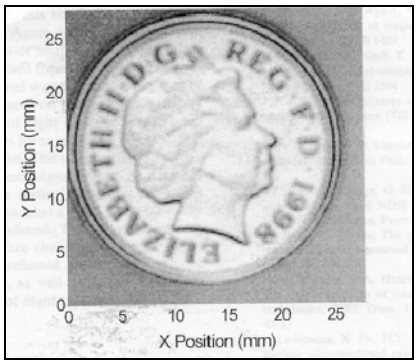

Fig 8-2. Coin image using gas-coupled Ultrasound [Robertson et al (2002)]

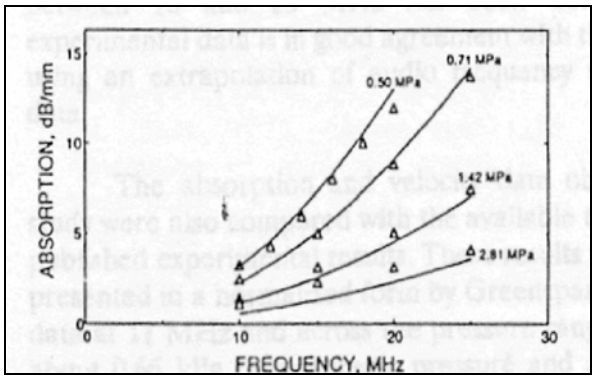

Fig 8-3. Attenuation as a function of pressure in nitrogen [Bond (1992)]

Electrical Impedance – capacitance measurements

Electrical impedance measurements have been successfully used in geophysics, seismology, engineering (NDT) and medicine. Conductive variations have been used to both characterize and image flow. Most recently an electrical impedance microscopy was reported by [Xiang and Gao (2002)]. The image shown in Fig. 8-4 is a response from a small reference notch and corresponding frequency/impedance data from a line scan across the same feature.

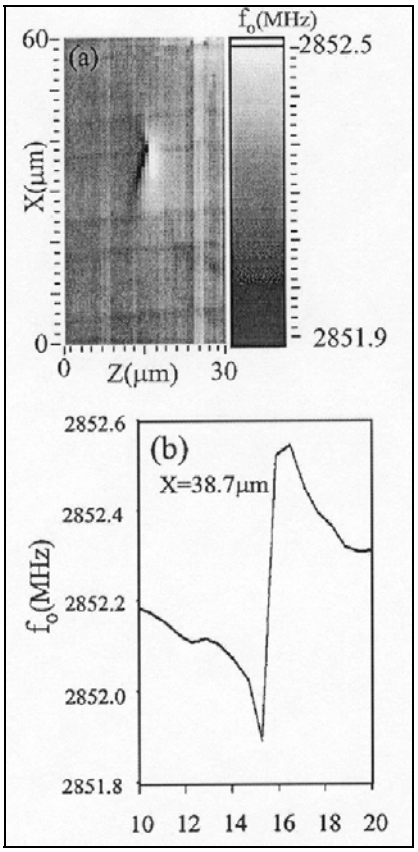

Fig. 8-4. Example of electrical impedance microscopy image [Xiang and Gao (2002)].

Ultrasound backscatter, scattering and diffuse field

Ultrasound has been implemented in several forms for the characterization of multi-phase systems [Tayebi et al (2001). Diffuse fields have been monitored in glass bead slurries [Weaver and Sachse (1995)] and there have been numerous studies of the response of "hard" targets that result in multiple-scattering of sound (ultrasound). This literature was recently reviewed by Tourin et. al. (2000). An example of the complex time domain and spectral response for diffuse transport of acoustic waves in glass beads in water is shown with the data given as Fig. 8-5 [Page et al (1999)]. The data shown as the RF data in Fig 8.5 are different from that used in most conventional ultrasound measurements in two respects: (i) the length of record considered—much longer time, and (ii) the nature of the signal. Convention pulse-echo or transit time measurements consider a primary pulse and consider amplitude change (attenuation) and arrival time. In diffuse-field/back-scatter, the signature results from multiple scattering and are at much lower levels than the primary pulses (see inset) where amplitude is ~ +- 1 compared with +- 0.002 in diffuse field.

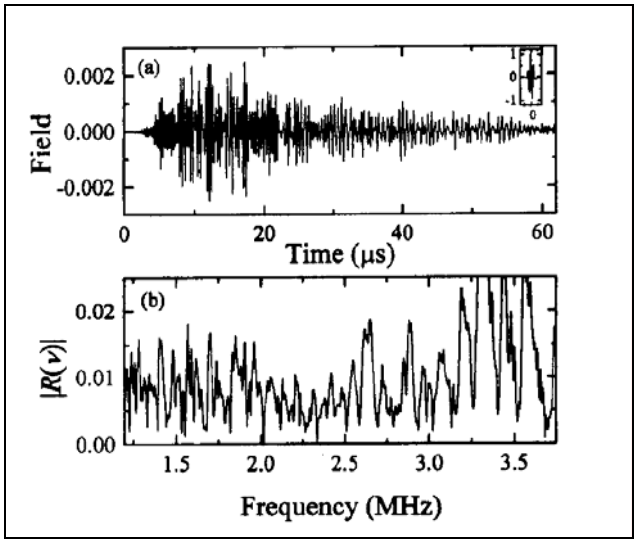

Fig. 8-5. Example of time domain and spectral response for diffuse transport of acoustic waves in glass beads in water [Page et al (1999)].

Tomography

There has been a growing interest in the application of tomography for the imaging of process chambers and flows [Williams and Beck (1995), Plaskowski et al (1995)]. Tomography can be performed in both reflection and transmission and the features of many implementations are shown in Fig. 8-6. The largest challenge in tomography is to get an adequate range of viewing angles, although some work has been successfully performed in reflection and involving limited viewing angles.

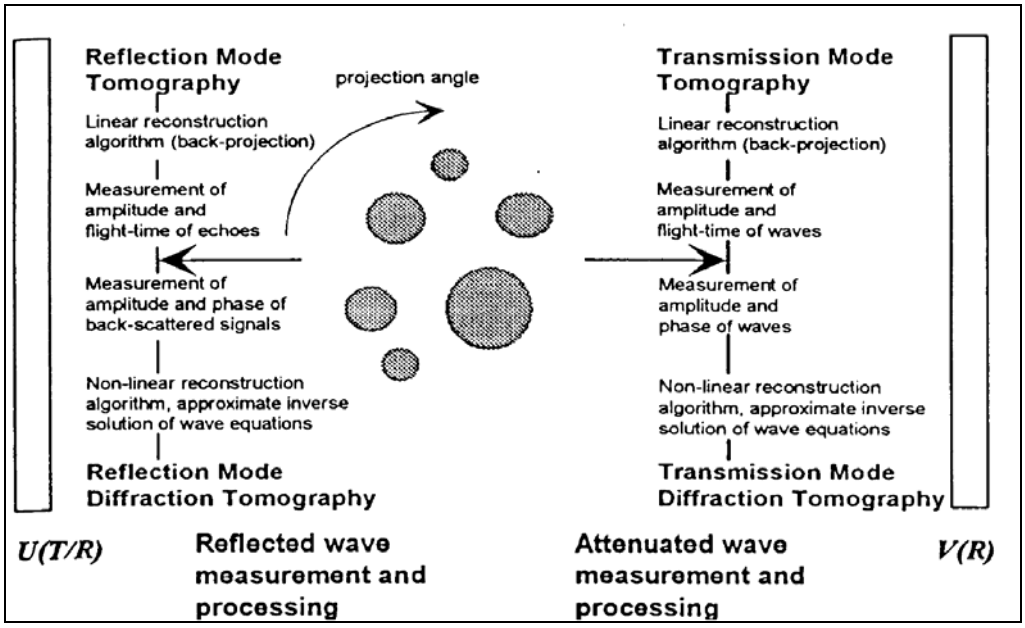

Fig. 8-6. Concepts involved in tomography [from Plaskowski et al (1995)].

Method assessment summary:

The technology assessment identified (i) transmission and (ii) back-scatter based ultrasound measurements to be the two approaches that could potentially be implemented using limited access in a high temperature system with a vessel that limits access. If successful, these ultrasound techniques do have the potential to be expanded and developed for implementation in a form of tomography.

In support of ultrasonic transmission and reflection measurements, the theory for multiple scattering for "hard" targets is quite well developed, including applications to gas-particle systems and implementations for fluidized beds.

The largest challenges are to be faced in achieving the required sensitivity, for example, to small mean-radius changes for a batch of particles and in achieving quantitative analysis for data interpretation that can potentially be complex.

(b) Preliminary results obtained with transmission ultrasound on a model coater.

Following from the identification of the potential for ultrasound based on a design for a unit at ORNL, a model coater was fabricated and used to investigate the effects of: (i) particle size and (ii) concentration on measured parameters. The coater-transducer system was also modified to (a) provide windows into which the transducers were inserted – which improved the signal/noise ratio by more than 20 dB and (b) copper coating was added to ground the vessel and eliminate static problems. The photograph in

Fig. 8-7 shows the fully functional model coater at PNNL after being modified.

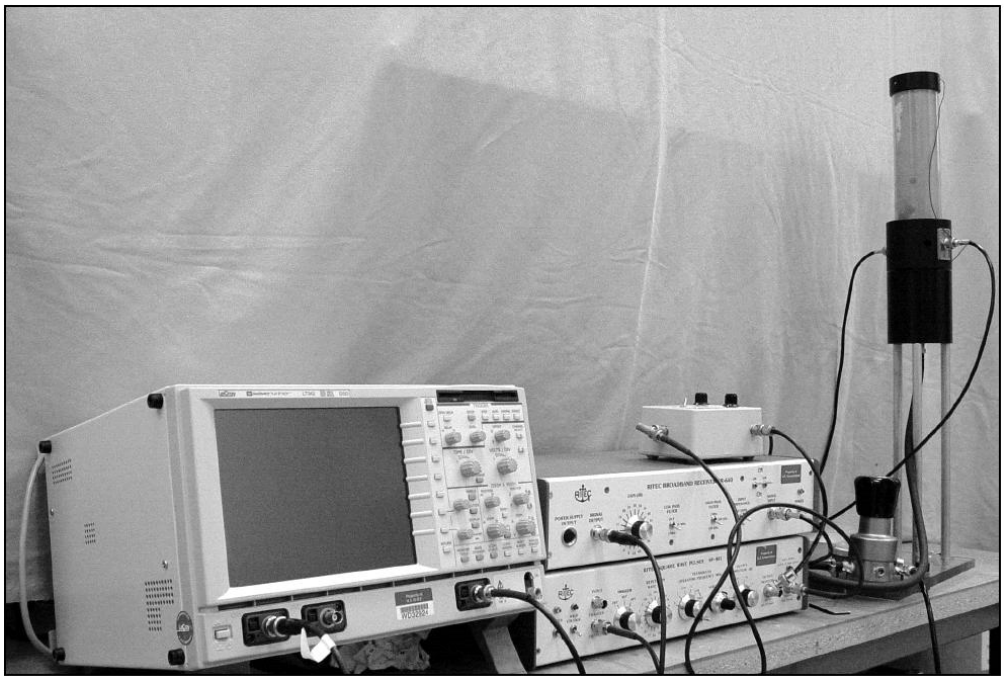

Fig. 8-7. Model coater and measurement system.

An example of changes in ultrasonic response seen with particle size and coater operating pressure are shown in Fig. 8.8. Example of change in measured time-of-flight with two different particle sizes and operating pressure, is shown in Fig. 8.9.

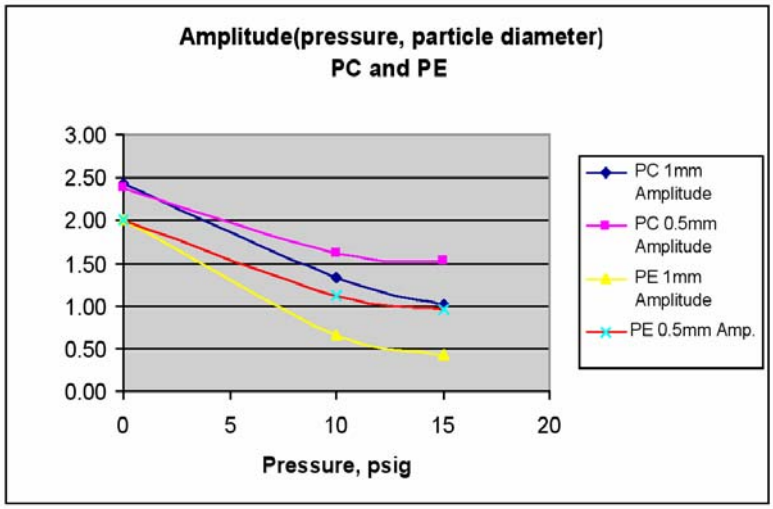

Fig. 8-8. Example of changes in normalized amplitude response seen with particle size and coater operating pressure.

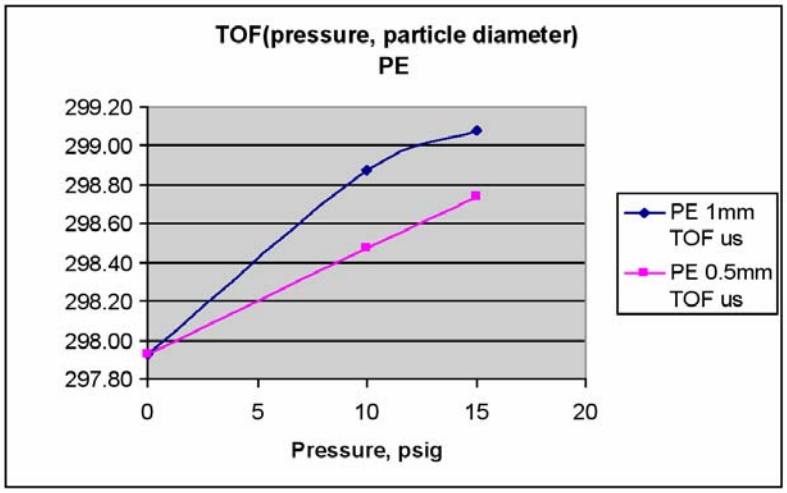

Fig. 8-9. Example of change in measured time of flight (microseconds) with two different particle sizes and operating pressures.

The preliminary data from this study and that in the literature do demonstrate that acoustic/ultrasonic methods have the potential to give on-line in-process thickness/diameter monitoring in the coater. It is now necessary to determine if this technique can provide the required sensitivity to particle parameters to monitor a process for an operating coater with TRISO particles during fabrication.

Next Steps

During FY 04 batches of coated particles will be obtained and trials performed that will seek to further investigate the capabilities for transmission ultrasound on-process measurement. In addition further work will be performed to review, and then potentially apply, models that have been used to predict ultrasound response in fluidized beds.

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Task 9. ESTABLISH STANDARD SIGNATURES AND FLAW LIBRARY

This task will develop a library of standard particles containing the full range of conditions known to degrade fuel performance, as well as particles deemed to result in optimal fuel performance. Multiple particles will be selected for each standard to enable statistical analyses. The standard particles will form a basis for developing and calibrating the NDE techniques for automated defect detection. For each NDE technique, a signature will be established for each particle in the flaw library.

In FY-03, considerable effort went into developing the NDE methods. The measurement results from the NDE methods showing the greatest potential for automated defect detection were evaluated using surrogate TRISO particles developed from another program that was in the early stages of developing the coating processes. The coating facilities at ORNL were unable to provide the specific needs of this task in FY-03. Therefore, large numbers of particles, having highly variable coating conditions, were sorted through to select out the few particles with defects representative of those considered to degrade fuel performance in a reactor. These particles were used predominately in finding trends between X-ray characterization and an electromagnetic measurement technique.

One of the outcomes from this effort was a clear definition of the type of particles that a flaw library must contain to fully develop, assess and calibrate the NDE methods. The matrix of coating runs and associated parameters to achieve the necessary standard particle library will be submitted to ORNL for review in October 2004. Even without particles coated specifically for this project, strong correlations were found between particle diameter and eddy current coil impedance, however, the multitude of undetermined parameters that could be varying among these early particles made it difficult to draw conclusions as to which parameters the NDE methods were most sensitive to and sort out the various effects. The coating runs to be developed early next year will simplify this problem by eliminating many of the simultaneously varying parameters and systematically focus on a limited parameter set.

One of the biggest challenges faced by this task (and other programs, as well) is particle characterization. To date there are a very limited number of methods available to facilitate characterization at ORNL's coating facility to verifying that specified coating parameters actually result in particles with particular properties. Calibration of the NDE methods being developed for automation requires independent calibration of each particle in the standard library. Micro-focus radiography and CT X-ray techniques were developed under Task 4a of this project for providing kernel size, layer-thickness and density distribution. Additional methods for characterizing coating layer porosity, density, and crystal structure are expected to become available at ORNL in FY-04.

PRESENTATIONS AND PUBLICATIONS:

Four presentations on work completed under this project were given this year, as well as the annual program review in Rockville, MD. The first presentation was to the 40th WANTO Meeting (Weapons Agencies Nondestructive Testing Organization) held at PNNL and titled "Micro-NDE", presented by Dr. Leonard Bond on April 22-24. The material presented at this meeting is generating interest within the NNSA community as techniques to assist NIF Target characterization.

A second presentation, titled "QA/QC for Advance Fuel Particle Production", was presented by Dr. Ronald Hockey at the annual ANS Meeting (American Nuclear Society) held on June 1–5, 2003 at the Town and Country Convention Center in San Diego, California. This presentation was a summary of work to date on using the NDE methods for fuel QA/QC under investigation by PNNL, GA, ISU and ORNL on this NERI project. The abstract is listed under: R. Hockey, L. Bond , M. Good and J. Gray, 2003. "QA/QC for Advanced Fuel Particle Production", Transactions Of The American Nuclear Society, Advances in Nuclear Fuel, Vol. 88, TANSAO 88 1–938 (2003), ISSN: 0003-018X. Presented at the 2003 ANS Annual Meeting, June 1–5, 2003; San Diego, California, pp. 412—413.

Dr. Salahuddin Ahmed presented the following two papers on the calculational modeling work of the TRISO fuel acoustic properties.

S. Ahmed and R. L. Hockey 2003, "Effects of Shape and Layer Defects on Resonance Frequencies of Spheres", presented in the NDE 2003 Conference, December 11-13, 2003, India.

S. Ahmed and P. D. Panetta 2003. "Effects of Layer Properties on the Ultrasonic Resonance of Composite Spheres", Review of Progress in Quantitative NDE KI Convention Center – Green Bay, Wisconsin, July 27 – August 1, 2003.

Financial Summary

Cumulative Cost Performance

	Oct	Nov	Dec	Jan	Feb	Mar	Apr	May	Jun	Jul	Aug	Sep
Cum Actual ($K)	$10.4	$21.7	$37.0	$63.1	$97.8	$155.3	$192.0	$233.9	$300.3	$350.6	$423.4	$540.1
Cum Budget ($K)	$35.6	$41.4	$124.2	$206.8	$267.7	$305.0	$372.8	$436.3	$484.1	$530.9	$546.4	$570.5

Annual Cost Performance:

Expenditures of $540.1K were incurred during FY03 of the $570.5K received in funding. Spending lagged in the first four months of the year while two subcontracts and non-disclosure agreements with Iowa State University and General Atomics were negotiated and established. Spending accelerated continuously over the next eight months peaking in September due to lagging subcontract invoicing and accruals of estimated cost for the last quarter.

During the second quarter funds were deobligated due to the closure of the DOE Oakland Operations Office at the end of FY03 and reinstated in the fourth quarter by DOE HQ.

A change request to carry over approximately $30K of FY03 scope and associated funding was requested and approved during the month of September.

Funding for FY04 arrived at PNNL during the fourth quarter allowing planning for FY04 and subcontract actions to be initiated.

Project Milestones:

Milestone & Deliverables	Planned Completion	Revised Completion	Actual Completion	Percent Complete
Place subcontract with ISU	Oct—02		Dec-02	100
Place subcontract with GA	Oct—02		Dec-02	100
Nondisclosure Agreements	Nov—02		Nov-02	100
Complete Initial Literature Review	Jan—03		Jan-03	100
Quality Index Defined FY-03	Jun—03		Jun-03	100
Particle Batch No. 1 & CT Received	Apr—03		Aug-03	100
Additional Particles & CT	Jun—03		Dec-03	100
Stationary NDE Method Development Complete	Sep—03	Dec-03	Jan-04	100

Standard Signature Flaw Library Complete	Apr—04	Jun-04		5
FY-03 Annual Report Complete	Sep—03	Mar-04		90
FY-03 Annual Report Complete	Oct—04			

The stationary NDE methods development and the standard signature flaw library tasks are behind schedule due to delays in obtaining surrogate particles with the coating properties necessary for testing and calibrating the NDE methods. Measurement data from the In-line NDE measurement methods will be necessary to develop the quality index. Particles appropriate for completing these milestones are expected to become available in the first quarter of FY-04, barring further delays in the coating development work at ORNL.

Appendices

ORNL delivered particles left over from the AGR program.

Table A1. ORNL supplied particles supplied September 9, 2003.

		Kernel			
Coating Run	**Cone ID**	**Material**	**Diameter**	**Charge In**	**Layers**
NT-	**(mm)**		**(µm)**	**(g)**	
52	25	ZrO2	500	10.008	Buffer + IPyC + SiC
64	25	ZrO2	500	10.009	Buffer + IPyC + SiC
74	50	ZrO2	500	54.5	Buffer + IPyC + SiC
75	50	ZrO2	500	54.5	Buffer + IPyC
AGR-08282003-1	50	ZrO2	500	54.5	Buffer
AGR-08282003-2	50	ZrO2	500	54.5	Buffer
AGR-08282003-3	50	ZrO2	500	54.5	Buffer
AGR-08292003-1	50	ZrO2	500	54.5	Buffer
AGR-09022003-1	50	ZrO2	500	54.5	Buffer
AGR-09032003-1	50	ZrO2	500	54.5	Buffer + IPyC
AGR-09042003-1	50	ZrO2	500	54.5	Buffer + IPyC
HfO2, Buffer only	50	HfO2	500	92.7	Buffer
HfO2, B + IPyC	50	HfO2	500	92.7	Buffer + IPyC

General Atomic's Deliverable No. 1: Derived From the report "Criteria for Developing Advanced QC Methods for Gas-Cooled Reactor Fuel Particle."

Table GA 1. Recommended Coated Particle Standards for QC Methods Development and Qualification

Key Property	Priority[1]	Potential Method	Particle Type Needed	Particle Standards Required[3]	Comments
Kernel stoichiometry	2		Kernel	UO_2 kernels, UCO kernels, and UC_2 kernels. Alternately, UCO kernels having different O/U and C/U ratios, if these ratios can be established in individual kernels	Availability of UC_2 kernels is doubtful. O/U and C/U may be determinable in individual kernels by x-ray diffraction
Missing (or thin[2]) buffer coating	1			Particles with nominal buffer density and the following buffer coating thickness: - no coating - 20 microns - 40 microns	Determine actual buffer coating thickness by radiography
Buffer coating density	3			TBD	Possible to characterize buffer void volume as a verification of both buffer coating thickness and density?
IPyC microstructure	1			Particles with high, moderate, and low anisotropy and nominal IPyC thickness and density: - IPyC deposited with CGR = 0.15 - IPyC deposited with CGR = 0.25 - IPyC deposited with CGR = 0.35	Characterize anisotropy with optical OAF measurement
IPyC thickness	2			Particles with low anisotropy and the following coating thickness: - 35 microns - 20 microns	35 micron standard is also one of the IPyC microstructure standards
IPyC density	3			TBD	
Missing or thin SiC	1			Particles with following SiC coating thickness: - 15 microns - 25 microns - 35 microns	Determine actual SiC coating thickness by radiography

Key Property	Priority[1]	Potential Method	Particle Type Needed	Particle Standards Required[3]	Comments
Cracks or pores penetrating the SiC layer	1			Particles with pores in SiC - Laser drilled surrogate particles - Particles with SiC deposited on an IPyC-coated substrate having a highly permeable IPyC - Particles with cracked SiC	Verify presence of pores using burn-leach test Create cracked SiC by applying a compressive load to particles
Lenticular flaws in SiC coating	3			Particles with small, medium, and large "gold spots"	Fabricate by running a SiC batch with excessive particle fluidization
Metallic impurities in SiC coating	1			Particles with metallic inclusions - At IPyC/SiC interface - At SiC/OPyC interface	Fabricate by introducing metallic impurities into surface of IPyC prior to SiC coating and into surface of SiC prior to OPyC coating
SiC microstructure	1			Particles with SiC deposited at: - 1400C (globular structure, α-SiC, and free Si) - 1550C (small grains, β-SiC) - 1750C (very large columnar grains, β-SiC, and free C)	Characterize particle batches by ceramography (structure) and Raman spectroscopy (SiC structure and presence of free Si and C)
OPyC microstructure	2			Particles with high, moderate, and low anisotropy and nominal OPyC thickness and density: - OPyC deposited with CGR = 0.15 - OPyC deposited with CGR = 0.25 - OPyC deposited with CGR = 0.35	Characterize anisotropy with optical OAF measurement
OPyC thickness	3			Particles with low OPyC anisotropy and the following coating thickness: - 40 microns - 20 microns	40 micron standard is also one of the OPyC microstructure standards
OPyC density	3			TBD	

[1]1 is highest priority and 3 is lowest
[2]A buffer coating is considered to be "thin" if the thickness is less than 20 microns
[3]Particle standards in bold type will be available from coating runs performed under the AGR Program. However, all of these particles will contain depleted or natural uranium kernels

General Atomic's Deliverable No. 2: A summary describing the TRISO particle coating process.

Table GA2. TRISO coating gases, coating rates, active coating gas fractions, and temperatures.

Coating	Diluent and Levitation Gas	Active Coating Gas	Mean Coating Rate (μm/min)[(a)]	Active Coating Gas Fraction[(b)] C/(C+L+D)	Nominal Coating Temperature[(c)]
Buffer	Ar, or Ar and He	C_2H_2	(d)	(d)	1250
IPyC	Ar	C_2H_2 and C_3H_6	≥3.0	0.25 – 0.35	1300
SiC	H_2	CH_3SiCl_3	≤0.33	0.012 – 0.021	1500
OPyC	Ar	C_2H_2 and C_3H_6	≥3.0	0.25 - 0.35	1300

Notes:

a) Mean coating thickness divided by coating deposition time.
b) C = active coating gas flow rate to coating zone (C_2H_2 + C_3H_6 for PyC coatings only), (CH_3SiCl_3 for SiC coatings only). L = levitation gas flow rate to coating zone (Ar for PyC coatings only), (H_2 for SiC coating only). D = diluent gas flow to coating zone (Ar for PyC coatings only), H_2 for SiC coating only).
c) Normal temperature in the active coating zone of particle bed.
d) Not defined.