Evaluating Quantitative 3-D Image Analysis as a Design Tool for Low Enriched Uranium Fuel Compacts for the Transient Reactor Test Facility: A preliminary study

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Abstract

3-D image analysis when combined with a non-destructive examination technique such as X-ray computed tomography (CT) provides a highly quantitative tool for the investigation of a material's structure. In this investigation 3-D image analysis and X-ray CT were combined to analyze the microstructure of a preliminary sub-sized fuel compact for the Transient Reactor Test Facility's low enriched uranium conversion program to assess the feasibility of the combined techniques for use in the optimization of the fuel compact fabrication process.

The quantitative image analysis focused on determining the size and spatial distribution of the surrogate fuel particles and the size, shape, and orientation of voids within the compact. Additionally, the maximum effect of microstructural features on heat transfer through the carbonaceous matrix of the preliminary compact was estimated.

The surrogate fuel particles occupied 0.8% of the compact by volume with a log-normal distribution of particle sizes with a mean diameter of 39 μm and a standard deviation of 16 μm . Roughly 39% of the particles had a diameter greater than the specified maximum particle size of 44 μm suggesting that the particles agglomerate during fabrication. The local volume fraction of particles also varies significantly within the compact although uniformities appear to be evenly dispersed throughout the analysed volume. The voids produced during fabrication were on average plate-like in nature with their major axis oriented perpendicular to the compaction direction of the compact. Finally, the microstructure, mainly the large preferentially oriented voids, may cause a small degree of anisotropy in the thermal diffusivity within the compact. $\frac{\alpha_{\parallel}}{\alpha_{\perp}}$, the ratio of thermal diffusivities parallel to and perpendicular to the compaction direction are expected to be no less than 0.95 with an upper bound of 1.

Keywords: 3-D image analysis, ZrO_2 surrogate fuel, LEU TREAT conversion, thermal diffusivity

1. Introduction

The push for new accident tolerant nuclear fuels and fuel designs has created a strong need for testing under extreme accident-like transient conditions. These new fuels and core materials need to be characterized under various levels of radiation exposure to ensure they will behave as designed in an extreme accident scenario. The desired reactor environment may be established using controlled transient testing where the nuclear fuel and core materials of interest are subjected to short bursts of intense high-power radiation inside a nuclear reactor core.

In response to the increased need for testing new nuclear fuels under transient conditions, the United States Department of Energy has authorized the reestablishment (restart) of the Transient Reactor Test Facility (TREAT) at Idaho National Laboratory [1]. The TREAT reactor first achieved criticality in 1959 and achieved more than 6,000 reactor startups before its operation was suspended in 1994. During its lifespan, TREAT was used as a tool for the design of durable nuclear fuels, establishment of performance limits, validation of design codes, and assisting in the definition of safety limits for new nuclear reactor core designs. During a transient, TREAT is capable of depositing high fission energies in test specimens without damage to the driver fuel.

In conjunction with the TREAT restart initiative, the National Nuclear Security Administration seeks to replace the existing highly enriched uranium (HEU) core with a low enriched uranium (LEU) core (i.e., with ^{235}U content less than 20% by weight) in order to reduce the risk presented by radiological materials [2]. The new LEU fuel, similar to the original HEU

fuel, will consist of a mixture of fine particles of UO_2 and graphite formed into a square-shaped columnar element with a metallic cladding (See Fig. 1 [3]).

The fundamental functions of the TREAT LEU design focused specifically on the fuel blocks and are listed below [4]. These provided the basis for exploring quantitative 3-D image analysis for fuel block non-destructive examination.

- Performance capabilities corresponding to TREAT's mission needs:
 - Proper heat conduction and adequate capacity for transient-generated energy deposited in the driver fuel (thermo-mechanical properties of fuel meat).
- Dimensional stability of fuel element assemblies (retain steady-state coolability and predictable nuclear configuration):
 - Fuel meat resistance to fractures driven by thermal shock, fission damage, and gas evolution
 - Adequate retention of gases evolved during irradiation and high-temperature service to prevent bulging or pillowing
 - Manageable thermal creep, elongation, swelling, and other environmentally driven structural behaviors.
- Chemical stability of fuel element assemblies:
 - Inertness or benign reactions between dissimilar materials (can to fuel block contact, fuel particle to graphite matrix, can to end caps).

The performance and dimensional stability of the TREAT LEU design are directly tied to three main components of the fuel microstructure, namely the dispersed UO_2 fuel particles, the graphitic matrix, and the voids within the material resulting from the fabrication process.

The fuel particle size and spatial distribution within the matrix are critical to fuel neutronics performance. Significant variations in the local size or spatial density of the fuel particles could lead to "hot" or "cold" spots within fuel blocks, which could result in significant local stress within the material. Over numerous cycles this stress could lead to failure within individual fuel blocks.

The inevitable formation of voids during the fuel fabrication will degrade the mechanical performance of the fuel blocks relative to the ideal block design without porosity. Voids, especially those with high aspect ratios, will act as stress concentrators. Over many thermal cycles of TREAT voids may become initiation sites for the propagation of cracks through the fuel blocks. Moreover, carbonaceous structures are prone to irradiation induced creep and dimensional change. Anisotropy in the porosity may lead to non-uniform overall dimensional change over time. Given these factors, it is important to understand the quantity, size, and shape of voids produced during the fabrication process.

Finally, the carbonaceous matrix is the primary path for removing heat from the fuel particles to the external environment. If the voids have preferred orientation within the compact, it may cause directional variations in the effective rate of heat transfer, but it also has the potential to cause significant variations in temperature if a large amount of transient-generated energy is deposited within the fuel over a short period of time. The temperature variations in turn could increase stress levels within the core.

These three microstructure components can be imaged non-destructively by means of micro X-ray computed tomography (μXCT) and quantified via 3-D image processing and subsequent image analysis. This study utilizes the non-destructive analysis of μXCT to provide preliminary insight into the effect of the fabrication process on fuel particles and void formation. For fuel

particles, the particle size distribution and spatial distribution is determined. Voids are analysed in terms of the size, shape, and orientation. In addition, the matrix is assessed for differences in the thermal diffusivity in the directions parallel and perpendicular to the compaction direction.

2. Experimental

2.1. Compact production

As part of the TREAT LEU conversion trade off study [2], two fabrication methods (namely, extrusion and compaction) were evaluated. Preliminary studies showed feasibility of both processes, but compaction was selected based on the significant advantage of the higher graphitic content (graphite to total carbon ratio) of 0.91 versus the 0.72 of the extrusion process. Babcock & Wilcox Nuclear Operations Group successfully demonstrated a mix, overcoat, and compaction method to produce sub-sized compacts with a nominal diameter of 1/2-in. diameter by 1-in. tall (Fig. 2). Figure 3 illustrates the fabrication process. The process starts with the jet milling (an alternate route would replace jet milling with high shear mixing) graphite, resin, curing agent, and surrogate ZrO_2 particles in the place of UO_2 . Initial engineering specification for the fuel particles require a particle size no larger than 44 μm . This corresponds to a 325 mesh sieve. The mixture is then granulated (overcoating) and vacuum dried before compaction. After compaction the fuel blocks are heat-treated to carburize the resin. The sub-sized compact used in this preliminary analysis was slowly heated to 950°C and held at temperature for 30 minutes before cutting furnace power and cooling to room temperature. As part of the compaction feasibility study performed, four different compaction lots were fabricated with varying parameters in a preliminary attempt to optimize them during fabrication scoping studies. A random compact from batch 13104-02 was chosen for this initial study.

2.2. μXCT Scanner

X-ray radiographs for computed tomography and 3-D reconstruction were acquired using a μXCT system located at Idaho National Laboratory. The source was a 130 kV micro-focus x-ray tube with an 8 μm nominal spot size. The detector was a Varian 2520V amorphous silicon flat panel with 127 μm pixel pitch and a 19.3 cm by 24.2 cm active area. A rotation stage between the source and detector provided precision rotation, enabling computed tomography. The source was operated at 130 kV and 48 μA with a 0.15 mm thick copper filter for acquisition of all projections acquired for this preliminary analysis.

The imaging system geometry for this study was determined through μXCT calibration using a calibration specimen (an array of ball bearings separated by 0.762 mm) and proprietary software provided by North Star Imaging. The source to detector distance (698.9 mm), source to rotation axis distance (40.6 mm), and rotation axis to detector distance (658.3 mm) were determined from the calibration. The resulting magnification was 17.22, yielding an effective pixel pitch of 7.4 μm . Projections were acquired every 0.5° for a total of 720 projections, and each projection was an average of three exposures. Tomographic reconstruction was performed using North Star Imaging proprietary reconstruction software. Fig. 4 shows a representative slice of the resulting reconstruction.

The reconstruction was largely free of obvious imaging artifacts, having no major beam hardening or ring artifacts. There was some rippling, which was more clearly visible outside the specimen in the void space. This artifact appeared as texture in the reconstruction where there was no such texture in the actual specimen. This artifact is inherent to CT reconstructions and can be minimized by increasing the frame integration time or increasing the number of projections. These ripples may manifest as a lower signal-to-noise ratio that can negatively

impact quantitative measurements derived from the gray level intensity of the reconstructed slices.

2.3. X-ray System Characterization

The effective spatial resolution of the x-ray imaging system was quantitatively determined using a modulation transfer function derived from an edge spread function [5]. The edge specimen used for this measurement was a 0.125 mm thick lead foil. The resulting image of the edge specimen was processed to produce a pre-sampled edge spread function. The line spread function was acquired by taking the discrete derivative of the edge spread function, and the modulation transfer function was calculated as the fast Fourier transform of the line spread function. The resulting modulation transfer function is shown in Fig. 5. The Nyquist frequency (f_{Ny}) for a 7.4 μm pixel pitch is 67.65 line pairs (lp) per millimeter. The spatial resolution of the imaging system at 10% contrast was 42.0 lp/mm (23.8 $\mu\text{m}/\text{lp}$) at 10% contrast. With image editing software, it was possible to distinguish features at contrasts as low as 2%, which gives an effective spatial resolution of ~ 52.5 lp/mm (19 $\mu\text{m}/\text{lp}$) according to Fig. 5. The effective spatial resolution was approximately equal to the reconstruction pixel pitch; therefore, the reconstruction pixel pitch was used as the effective voxel resolution.

2.4. Image Processing

To extract quantitative data from the 3-D volume, the voxels corresponding to particles and voids must be accurately identified. In the initial dataset (Fig. 4), significant overlap in intensity existed between the voxels corresponding to the graphitic matrix material and voids within the compact. The following pre-processing steps were taken to minimize this overlap and provide a better representation of the microstructure upon image segmentation. First, as shown in Fig. 6a and 6b, the initial data volume was cropped to the largest possible parallelepiped volume with its lengths in the x and y direction equal to one another. Next, a local adaptive wiener filter was used to filter out noise in the data set. A wiener filter with a 7×7 voxel kernel was used to remove a significant portion of the noise within the image without artificially blurring features. The result of applying a wiener filter to Fig. 6b is shown in Fig. 6c. To further distinguish the matrix material from the voids, an h-minima transform was used. The h-minima transform suppresses all local minima with a value less than or equal to a given threshold depth, h. A comparison between Fig. 6c and 6d showed that many small local minima corresponding to noise within the matrix material were removed after using the h-minima transform. As a result, voids were more easily distinguishable in terms of intensity.

To mitigate user bias in selecting threshold parameters, an automated global threshold algorithm known as Otsu's method was used [6]. Otsu's method selects one or more threshold values (the number of thresholds is set by the user) by finding the threshold(s) that maximize the variance of the intensity within the data set. Two thresholds were used in this preliminary study, to distinguish the surrogate particles, matrix, and voids. Values greater than $\sim 65\%$ of the maximum voxel intensity represented the fuel surrogate ZrO_2 particles within the compact. Voids within the compact were well represented by voxels with intensity less than $\sim 37\%$. Voxel intensities between the two threshold values were considered matrix material. Images corresponding to Fig. 6d with the isolated ZrO_2 and voids are shown in Fig. 7a and 7c, respectively. Prior to analysis of the isolated voids, the void volume was morphologically closed with a $3 \times 3 \times 3$ voxel kernel to remove obvious artifacts. Volumetric representations of the ZrO_2 particles and voids are shown in Fig. 7b and 7d, respectively.

2.5. Algorithms and Mathematical Operations

Several mathematical operations were used to extract quantitative results from the data volume. Many of the concepts and methods used can be found in digital image processing texts [7–10]. For this preliminary 3-D analysis the methodology was simply expanded to include a third dimension.. In section 3.1, volume fractions were retrieved from the respective binary volumes by counting the number of voxels representing each component of the microstructure and dividing by the total analysis volume.

The fuel surrogate analysis, section 3.2, used a connected component algorithm to group individual voxels into particles. Briefly, a connective component algorithm groups connected voxels based on user-specified connectivity rules into objects. The output is typically in a form suitable for analysis of individual features isolated within the data.

Additionally, a 3-D discrete correlation of the form

$$w(x, y, z) \star f(x, y, z) = \sum_{s=-a}^a \sum_{t=-b}^b \sum_{u=-c}^c w(s, t, u) f(x + s, y + t, z + u) \quad (1)$$

was used where w represents a 3-D kernel or filter and f represents the data volume to be analyzed.

$$a = (m - 1)/2 \quad (1a)$$

$$b = (n - 1)/2 \quad (1b)$$

$$c = (p - 1)/2 \quad (1c)$$

and m , n , and p represent the dimension of w . For notational and computational convenience, m , n , and p are always odd integers. Allowing w to equal a 3-D matrix of ones, the discrete correlation effectively counts the number of voxels equal to 1 within f over a region specified by w . In section 3.2 the discrete correlation was used to determine the local volume fraction of particles within the local volume w .

Section 3.3, Void Analysis, utilized a connected components algorithm as previously described for surrogate particles in section 3.2 to group voxels into individual voids. A Hotelling transform was applied to each void to determine its size shape and orientation within the compact volume. A Hotelling transform creates a covariance matrix from the set of position vectors of all voxels within the void of interest. The three Eigen vectors of the respective covariance matrix represent the three principle axes of each void and thus the orientation with respect to the original coordinate system. The Eigen values of the covariance matrix are directly proportional to the length of each principle void axis. The Eigen values can be used as a quantitative measure of void size and shape. A 3-D Hotelling transform inherently assumes an ellipsoidal object.

Finally, in section 3.4 geodesic distance transforms were used to assess the maximum possible effect of the ZrO_2 particles and voids on bulk thermal diffusivity along directions parallel and perpendicular the compaction axis of the compact.. To do this, geodesic distance transforms were calculated along the two directions of interest. From the geodesic distance transforms the shortest path between opposite faces that passes through a given voxel was determined. This path was not allowed to pass through voxels that represented surrogate particles or voids. A multistencil fast marching method was used to calculate the geodesic distance transforms. The correlation between path length and thermal diffusivity is discussed in section 3.4.

3. Quantitative Image Analysis Results & Discussion

3.1. Volumetric Analysis

Using the binary volumes of the isolated surrogate particles and voids, estimates of the volumetric fractions were determined as discussed in section 2.5. The ZrO₂ surrogate comprised roughly 0.8% of the total volume, voids occupied approximately 2.2%, and the graphitic compact material by volumetric conservation represented the remaining 97%. In future compacts with UO₂, this analysis may be suitable for ensuring that the compact carbon to uranium ratio is correct.

3.2. Fuel Surrogate Analysis

Fig. 8 is a histogram of the ZrO₂ particle diameter plotted on a log scale (natural logarithm). The diameter was determined by measuring the volume of each particle and assuming a spherical geometry. The nearly “bell” shaped curve, on a log-scale, suggested the distribution was approximately log-normal. If the log-normal probability density function was defined as

$$P(x) = \frac{1}{S\sqrt{2\pi}x} e^{-(\ln(x)-M)^2/(2S^2)} \quad (2)$$

a non-linear fit of the particles yielded 3.58 and 0.40 for M and S , respectively. From these fitted values, the mean diameter, standard deviation, skewness, and kurtosis of the distribution were 38.9 μm , 16 μm , 1.3, and 3.2. Particles with diameters less than 25 μm were excluded from the log-normal fit to avoid the effects of discrete binning near the limits of the μXCT system resolution.

In Fig. 8, the first four occupied bins represent particles 2, 3, 4, and 5 voxels in size. As these particles are relatively close to the reconstructed resolution of the compact (7.4 μm) their size is most likely highly sensitive to small variations in the selected threshold. While the effect of noise was mitigated significantly by filtering the volume prior to analysis, it is likely that the small particles are also artificially high due to incorrect segmentation of a minute fraction of the matrix material. As the particle size increases, the accuracy of the diameter increases significantly.

A key observation from examination of Fig. 8 is the maximum particle size is greater than the engineering design limit of 44 μm . Roughly 39% of particles have a diameter greater than the design limit. Ignoring extreme outliers, the maximum particle diameter observed was 123 μm . To ensure the ZrO₂ powder met the engineering specification prior to fabrication, the powder was sifted with a 325 mesh sieve (nominal sieve opening of 44 μm). While sieves alone cannot finely control particle size, it is unrealistic to assume that nearly 40% of the particles passing through the sieve had a diameter significantly greater than the specified design. Consequently, a significant amount of agglomeration must occur during component mixing. Given this preliminary finding, work is currently underway to mitigate particle agglomeration via the introduction of a wetting agent during the mixing process.

To analyze the spatial distribution of particles within the compact, Eq. (1) was used. $w(x, y, z)$ was selected to be an isotropic 3-D matrix with all elements possessing a value of one. The matrix dimensions of $45 \times 45 \times 45$ voxels was used for the analysis. This kernel had an equivalent volume of nearly $3.7 \times 10^{-1} \text{ mm}^3$. The correlation was applied to every data point within the compact and normalized to yield the local volume % of surrogate particles. A random sample of 5×10^6 local volume fraction measurements from the data volume was used to produce the histogram in Fig. 9. The histogram was truncated to show more detail near the peak of approximately 0.5% ZrO₂ by volume. The maximum value from the random sample was ~17% ZrO₂ by volume.

The maximum likelihood value $\sim 0.5\%$ was skewed left of the bulk average of 0.8% ZrO_2 in section 3.1. This result suggested an appreciable degree of non-uniformity exists within compact and many regions exist with volume fractions significantly greater than the expected value.

Fig. 10 was generated to visually assess the distribution of the lowest and highest volume fractions. The lowest density regions, shown in red, were relatively small and were frequently observed throughout the volume. These regions were likely found near the centers of voids. The low-density regions corresponded to a volumetric fraction of $\text{ZrO}_2 \leq 0.03\%$. The high-density regions corresponded to zirconia volumetric fractions greater than or equal to 3% . These regions, shown in green, were significantly larger than the low-density regions and visually supported high-density non-uniformities in the form of high local number densities or particle agglomerations. While non-uniform, these globular high-density regions appeared evenly dispersed throughout the volume.

Particle agglomeration and spatial non-uniformities must be kept to a minimum to ensure the fuel behaves properly. The specification for maximum particle size was set based on initial calculations and estimates of the matrix thermal diffusivity to minimize the likelihood of melting fuel particles. As particle size increases, the contact area per unit volume of fuel decreases resulting in a higher average temperature within the particle. Even if all particles were to meet the design limit for particle size, high local densities of fuel particles could essentially produce the same effect by increasing the local temperature of the matrix and therefore decreasing the overall driving force for heat transfer away from the fuel particles. A uniform spatial distribution is also required to minimize hot and cold regions within the fuel blocks. Extremes in temperature will not only affect the neutronic performance, but may also over time lead to cracking and ultimately mechanical failure of a fuel block.

3.3. Void Analysis

The Hotelling transform, as briefly described in section 2.5, was used to ascertain the orientation, shape, and size of each void. As seen in Fig. 7d, a majority of voids were oriented parallel to the radial direction (perpendicular to compaction direction) of the compact. As shown in Fig. 11a, this corresponds to a large fraction of voids oriented with their primary and secondary axis perpendicular to the compaction direction. Consequently, by geometric definition of an ellipsoid, the tertiary axis is perpendicular to the plane made by the primary and secondary axis and was nearly parallel to the compaction direction in this preliminary investigation. The inset of Fig. 11a is an angular histogram plot of the azimuth angle of the primary void axis. The inset figure shows no statistically significant preferential orientation of the primary void axis. No preferred orientation was observed for the azimuth angles of any axis. There are two plausible explanations for the formation of the observed voids with their preferred alignment. The first explanation is the compaction process deforms the granulated matrix and the subsequent heat treatment is insufficient for the complete healing of the deformation. The second explanation is the volatiles formed from resin carbonization escape preferentially along the direction perpendicular to the compaction.

Fig. 11b plots the two aspect ratios of each void in a 3-D histogram. The primary aspect ratio was defined as the ratio of the secondary to primary axis length, and the secondary aspect ratio was similarly defined as the ratio of the tertiary to primary axis length. The average void shape, shown in the inset of Fig. 11b, had primary and secondary aspect ratios of 0.63 and 0.14 , respectively. On average, the voids within the compact were plate-like in nature.

Finally, Fig. 11c shows the estimated primary axis length of each void. The histogram of void length appeared to be bimodal with peaks at $\sim 150\ \mu\text{m}$ and $\sim 300\ \mu\text{m}$. Although a majority of

these voids fell near these two peaks, a number of them had large macroscopic dimension. The 13 largest voids have lengths greater than 1 mm (Fig. 12). These large flaws combined with their preferred alignment along the radial direction of the compact may play a significant role in the mechanical strength of the compact.

3.4. Estimation of Thermal Diffusivity Variation

With the preferred orientation and the average plate-like anisotropic shape of voids within the compact, the heat transfer rate through the compact may vary to some degree with compact orientation. Graphitic materials have high thermal diffusivity values along $[i\ j\ 0]$ crystallographic directions while along the $[0\ 0\ 1]$ direction thermal diffusivity is orders of magnitude slower. Graphite powder was only a small fraction of the mixture used for compaction. It accounted for an estimated 0.43% of the initial carbon mass, thus a majority of the compact matrix and resin material initially exists as large polycyclic aromatic hydrocarbons. The heat treatment at 950°C is not sufficient to graphitize the carbonaceous material. Rather hydrogen, oxygen, nitrogen, and other organic impurities are removed from the carbon backbone and the remaining carbon from the polycyclic aromatics essentially forms small and likely randomly aligned graphene sheets. The size of these sheets will vary depending on the precursor but likely contain ten to twenty six member carbon rings. In the production of graphite, the green body is heat treated to temperatures between 2500°C and 3000°C and the resulting polycrystalline graphite often has individual crystallite sizes on the order of 1 nm. Given the above explanation, it is a reasonable to assume that the thermal diffusivity of the matrix material is isotropic within the effective spatial resolution of the μ XCT scan.

The matrix material should be the primary path for heat transfer through the compact and is expected to have a thermal diffusivity significantly higher than ZrO_2 or voids. Assuming conduction through the ZrO_2 particles and voids is infinitely slower (perfect insulators) than conduction through the carbonized matrix and the matrix is indeed thermally isotropic on the scale of interest, an estimate of the maximum effect of microstructure anisotropy on thermal diffusivity can be determined.

The effective thermal diffusivity through a composite should be adequately described by

$$\alpha_{\text{eff}} = \frac{\alpha\phi}{\tau^2} \quad (3)$$

which is based on an analogous expression for mass diffusion through porosity [11]. In equation 3, α is the thermal diffusivity of the “pure” matrix material, ϕ is the volumetric fraction of composite represented by matrix material, and τ is the mean geodesic tortuosity of the conduction path through the matrix of the composite. The geodesic tortuosity can be defined as the geodesic path length between two points normalized to their Euclidean distance.

Fig. 13 displays a geodesic tortuosity map of the compact volume taken parallel to the compaction direction from three different viewpoints. From Fig. 13b and 13c especially, it is apparent that the shortest paths through the compact volume occur where there are few voids in the direct path. When voids are encountered, heat must flow around it and this increases the effective path length for heat transfer. When heat flows perpendicular to the compact direction, heat must still flow around voids, but the “cross-sectional” profile of the voids are significantly smaller in the perpendicular direction due to the plate like shape and preferential alignment of the voids.

Since the matrix thermal diffusivity is assumed isotropic and the volume fraction of matrix is independent of direction, the ratio of thermal diffusivities parallel and perpendicular to the compaction direction should be related to the respective geodesic tortuosities as follows.

$$\frac{\alpha_{\parallel}}{\alpha_{\perp}} = \frac{\alpha\phi/\tau_{\parallel}^2}{\alpha\phi/\tau_{\perp}^2} = \left(\frac{\tau_{\perp}}{\tau_{\parallel}}\right)^2 \quad (4)$$

The mean tortuous path along the compaction direction was equal to 1.0263 times the Euclidean distance, whereas the tortuosity in the radial plane was 1.0039. Using Eq. (4), the ratio of thermal diffusivities in the two respective directions should be no less than 0.957, with an upper bound of 1. Preliminary measurements of thermal diffusivity on a different batch of compacts during the initial trade study [2] suggested that, on average, compacts would have a thermal diffusivity parallel to the compaction direction of $11.38 \frac{mm^2}{s}$. Applying Eq. (4), the maximum thermal diffusivity that may occur in the perpendicular radial plane is $11.89 \frac{mm^2}{s}$, leading to a maximum difference of $0.51 \frac{mm^2}{s}$. As the fuel blocks for TREAT will primarily conduct heat perpendicular to the long axis of the fuel element assembly, this difference in the parallel and perpendicular directions will not have a significant effect on heat transfer.

4. Error and Limitations of Preliminary 3-D Quantitative Analysis

3-D quantitative analysis of geometric features is an extremely powerful characterization technique because it avoids many of the limiting assumptions that must be made in a similar 2-D analysis. In any image analysis, whether 2-D or 3-D, where feature size is represented by a small number of pixels or voxels, the analysis is very much limited by the feature size relative to the measurement resolution and prone to significant error as feature size approaches the imaging resolution. In this preliminary analysis, the ZrO_2 particle size was prone to error as a consequence of the reconstructed resolution. This affect was clearly visible on the left side of Fig. 8 as the first three occupied bins represent objects that differ in size by only one voxel. Additionally, many of the peaks to the left of the log-mean average, M , corresponding to a diameter of $38.9 \mu m$ were artificially high due to a similar affect. Particles represented by an effective diameter of $\sim 40 \mu m$ that were inaccurately binned by just one effective voxel length in diameter could have a diameter measurement in error by as much as 20%. For a value of 3.92 this decreased to approximately 12% and near 4.16 this relative error decreased to less than 10%. Taking this into consideration, the general shape of the curve was accurate, but the actual location of the log-mean average may be further to the left or right depending on thresholding error.

The local density distribution was not strongly influenced by error in the actual particle threshold due to the volumetric kernel size used for the analysis. Furthermore, segmentation error did not contribute significantly to error in the void dimensional measurements as voids were significantly larger than the effective voxel resolution.

5. Conclusions and Recommendations

A preliminary study of image processing and analysis has been performed on a single surrogate fuel compact to assess its feasibility as a quantitative design tool for the TREAT LEU

conversion fuel design. The microstructure was imaged with a μ XCT system and extracted features were quantitatively assessed in 3-D.

The size and spatial distribution of ZrO_2 surrogate fuel particles were determined to assess potential impact of fabrication on neutronic properties of the fuel. The particle size distribution was log-normal in nature with a mean particle diameter of $38.9\text{ }\mu\text{m}$ and a standard deviation of $16\text{ }\mu\text{m}$. Approximately 39% of zirconia particles isolated from the data volume had effective diameters greater than the maximum particle size of $44\text{ }\mu\text{m}$. These larger particles were likely particle agglomerates that formed during the mixing process prior to compaction.

The spatial distribution of surrogate particles peaked at approximately 0.5% ZrO_2 by volume, whereas the overall volume fraction was 0.8%. This along with visualization of voxels which make up the lowest and highest 0.25% volume fractions suggested a significant amount of high-density regions within the compact. Further process optimization must mitigate this effect to provide uniform neutronic performance throughout the fuel compact.

To more accurately assess the particle size in future work, the number of projections as well as the average number of frames per projection could be increased to increase the signal-to-noise ratio and more accurately resolve fuel particles within compacts. If future specimen are scanned at a significantly higher resolution, additional characterization of suspected agglomerations could be made to identify the number and size of each particle in an agglomerate.

These preliminary measurements are important to consider in the optimization of the fabrication method. A maximum particle size of $44\text{ }\mu\text{m}$ was set to achieve a desired neutronic performance from the fuel assembly while maintaining the fuel particle temperature significantly below its melting point. Agglomerations will lead to temperatures above those predicted in initial calculations and potentially cause the particles to melt at conditions thought to be within the safe operating conditions of TREAT. Similarly, regions with large volume fractions of fuel particles may cause increased particle temperatures, as the local matrix would be required to deal with a larger heat load.

The preliminary analysis has led to the introduction of a wetting agent during the mixing step of fabrication to mitigate agglomeration. Additionally, modelling efforts have begun to determine what degree of agglomeration and spatial non-uniformity can be tolerated within the fuel blocks without compromising the range of safe operation.

To assess voids within the sub-sized compacts a Hotelling transform was used to determine the shape, length, and orientation of voids. The vast majority of voids were plate-like in nature with primary and secondary aspect ratios of 0.63 and 0.14 (Inset Fig. 11b). Voids were preferentially aligned perpendicular to the compaction direction of the compact. The 13 largest voids had a major axis length greater than 1 mm. These dimensions are quite significant at nearly 10% of the compact diameter. Further analysis is needed to determine if these large voids are correlated to some characteristic length within the existing microstructure or the voids will increase in dimension as the processing is scaled up to fuel block production.

Finally, the effect of void anisotropy on the effective thermal diffusivity parallel and perpendicular to the compaction direction was assessed using geodesic distance transforms. The value measured for the ratio of thermal diffusivities in the two directions is a lower bound based on the assumptions that heat is transferred via conduction alone and the particles and voids within the structure are perfect insulators. The ratio measured comparing the two directions was approximately 0.957. Using preliminary measurements of thermal diffusivity of similar compacts, the absolute difference in thermal diffusivity would be no greater than $0.51\frac{\text{mm}^2}{\text{s}}$ at

100°C. This difference would not be expected to cause significant spatial variations in temperature within the fuel block design.

Based on these preliminary quantitative results, 3-D image analysis appears to be an excellent tool for optimization of the fabrication process as it is able to non-destructively interrogate and quantify microstructural features linked to the fuels ultimate performance. This method will be further examined in a controlled study with a larger population of compact samples with direct measurements of properties like thermal diffusivity and compressive strength in order to draw correlations between the microstructure and measured physical properties.

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References

- [1] P.B. Lyons, PBL-12-2-2010, December 3, 2010.
- [2] I.J. van Rooyen, N.E. Woolstenhulme, R.K. Jamison, E. Luther, and L.N. Valenti, INL/LTD-14-31704, April 2014.
- [3] I.J. van Rooyen, S.R. Morrell, R.K. Jamison, A.L. Crawford, H.T. Hartman III, E.P. Luther, A.E. Wright, D.C. Kontogeogakos, D.D. Papadimas, H.M. Connaway, and G. Yesilyurt, Paper INL/CON-14-33130, *RERTR 2014 – 35th International meeting on Reduced Enrichment for research and Test Reactors*, October 12–16, 2014, IAEA Vienna International Center, Vienna, Austria.
- [4] I.J. van Rooyen, D. Scates, R. Smith, R.K. Jamison, E. Luther, A. Crawford, T. Lister, and H. Hartman, PLN-4807, September 2014.
- [5] E. Samei, E. Buhr, P. Granfors, D. Vandenbroucke, and X. Wang, *Phys. Med. Biol.* 50, pp. 3613–3625 (2005).
- [6] N. Otsu, *IEEE Trans Syst Man Cybern.*, 9(1), pp. 62-66 (1979).
- [7] R.C. Gonzalez and R.E. Woods. *Digital Image Processing*. 3rd ed. Upper Saddle River, N.J.: Prentice Hall, 2008.
- [8] P. Soille, *Morphological Image Analysis: Principles and Applications*. 2nd ed. Berlin: Springer, 2003.
- [9] J. Serra, *Image Analysis and Mathematical Morphology*. Orlando, Florida: Academic Press, 1988.
- [10] J. Serra, *Image Analysis and Mathematical Morphology - Volume 2: Theoretical Advances*. London: Academic Press, 1988.
- [11] C.J. Gommers, A.J. Bons, S. Blacher, J.H. Dunsmuir, and A.H. Tsou, *AIChE Journal*, 55(8), pp. 2000–2012 (2009).

Fig. 1. Schematic presentation of the main characteristics of the (a) original and (b) upgrade HEU and the (c) draft conceptual LEU fuel element assembly design [3].

Fig. 2. An example of a sub-sized compact. The compact is $\sim 1/2$ -in. diameter by ~ 1 -in. tall [2].

Fig. 3. Compaction process flow chart [2].

Fig. 4. Representative CT reconstruction slice of compact (specimen 13104-02). The contrast has been enhanced for the visual identification of image artifacts.

Fig. 5. Effective spatial resolution of the x-ray imaging system used in this study. The calculation of effective spatial resolution was based upon a single projection.

Fig. 6. The brightness of each image within this figure was artificially enhanced by 40% to increase the visibility of features for the reader. Each image is taken from slice 790 within the image stack. Fig. 6a is a slice taken from the center of the compact reconstruction. Fig. 6b is cropped from the original slice in Fig. 6a. Fig. 6c shows the filtration of Fig. 6b with a 7×7 voxel kernel wiener filter. Fig. 6d shows the application a 3-D h-minima transform to Fig. 6c.

Fig. 7. Particles and voids isolated after pre-processing of the raw CT reconstruction. Fig. 7a shows the isolated particles, highlighted in blue, from Fig. 6b. Fig. 7b shows a small subset of particles within the 3-D volume. The width of the 3-D volume as shown is approximately 12.7 mm. The color corresponds logarithmically with particle size, with blue particles being the smallest (and most abundant), followed by green and orange. The red particles represent the largest and most infrequently observed particles. Fig. 7c shows voids, highlighted in red, from figure 6d. Fig. 7d is a 3-D rendering of all voids isolated within the 3-D volume. The width of the 3-D cross-section as shown in figure 7d is nearly 9 mm from the left to right side of the volume. The height of the volumes in figures 7b and 7d are approximately 11 mm.

Fig. 8. A histogram representing the distribution of ZrO_2 particle size within the analyzed compact.

Fig. 9. A histogram representing local volumetric fraction of zirconia within the fuel compact. The histogram was constructed from a random sample of five million voxels from the volumetric data set which contains more than 2.99 billion voxels. Each local measurement is has a sample volume of $333 \times 333 \times 333 \text{ } \mu\text{m}^3$.

Fig. 10. A visual representation of the lowest 0.25% and highest 0.25% percentiles of the local volume fraction analysis in Fig. 9. The red objects represent the lowest density regions $\leq 0.03\%$ zirconia by volume. The green objects represent the highest density regions within the compact with $\geq 3\%$ zirconia by volume. Fig. 10a shows the entire analyzed volume whereas Fig. 10b shows a closer view of the interior. The volume shown in figure 10a is approximately $9 \times 9 \times 11$ mm. The blue arrow at the top left of the figure is aligned with the volumes longest dimension.

Fig. 11. A visual representation the orientation, shape, and size analysis of voids within the fuel compact. Fig. 11a shows the elevation angle of the three principle axes. The inset angular histogram plot is the azimuth angle for the primary axis. Fig. 11b illustrates the shape distribution of voids within the compact. The average void shape is represented by the inset ellipsoid. Fig. 11c represents the distribution of void lengths within the compact. Fig. 11c tabulates only the primary axis lengths.

Fig.12. The 13 largest voids within the analyzed volume of the compact are shown. All voids shown have at least one dimension greater than 1 mm in length. The volume shown in is approximately $9 \times 9 \times 11$ mm. The blue arrow is aligned with the volumes longest dimension.

Fig. 13. The geodesic tortuosity taken along the compaction direction. Fig. 13a shows a map of tortuosity values taken from the same slice shown in Fig. 6d, which is perpendicular to the compaction direction (see Fig. 6 and 7). The color map bar correlates the image color to tortuosity value. Fig. 13b maps the tortuosity in a plane parallel to the compaction direction. The color map bar correlates the image color to tortuosity value. Fig. 13c is a 3-D representation of 1/4 of the entire compact volume analyzed. The voids and ZrO_2 particles are represented by red. Blue vertical lines represent the shortest pathways through the compact, followed by yellow and green regions. Dark “reddish-orange” paths seen in close proximity to large voids represent the longest path through the compact with a tortuosity near 1.09. The volume shown in figure 13c is approximately $4.5 \times 4.5 \times 11$ mm.