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Particle Homogeneity Analysis for Dispersion Target using X-ray CT

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1. Introduction

Mo-99 medical radioisotope has been mainly extracted from high enriched uranium (HEU) targets irradiated in the research reactors [1]. But after the non-proliferation policy claims to minimize the civilian HEU usage, the development of low enriched uranium (LEU) targets has been pursued [2]. Reduced isotope production efficiency of LEU target should be improved to resolve isotope shortage problem [3], preferably without increasing the amount of used targets, which leads to radioactive waste increase. Thus, it is necessary to develop LEU target with high uranium density [4].

In Korea, U-Al dispersion plates up to 9.0 gU/cm^3 density were fabricated based on centrifugal atomization technique [5], and the metallurgical reaction between U-Al during fabrication process has been investigated [6]. Interaction layers consisting of intermetallic compounds (UAl_x) were formed during the fabrication process, and lower density of the interaction layer leads to a volumetric increase, resulting in degradation of thermal properties of dispersion plate. Larger uranium particle sizes resolves interaction layer problem, but on the other hand, it causes uranium particle aggregation problem [7]. In order to obtain the homogeneous particle distribution, optimized fabrication conditions should be set by analyzing the internal microstructure of the dispersion plates.

X-ray CT technique has advantage of 1) obtaining entire internal information 2) without damaging the fabricated plates [8]. In this study, particle distribution of stainless steel – aluminum dispersion plates was analyzed to investigate the applicability of the X-ray CT technique on the microstructure analysis of high density LEU targets with coarse fuel particles.

2. Methodology Construction and Procedure

In order to obtain a desirable particle distribution, internal particle homogeneity should be evaluated with changing the fabrication conditions that affects the particle aggregation. Surrogate material for uranium fuel particles was selected and samples were prepared according to the conventional picture-frame method [9, 10]. The applicability of the X-ray CT technique was confirmed and a methodology estimating the internal particle homogeneity from CT data was established. The whole procedure is as follows. 1) Sample fabrication, 2) CT microstructure observation, 3) Estimation of internal particle homogeneity.

2.1 Materials and Fabrication

Because type 304L Stainless steel has yield strength similar to that of uranium, it is expected to behave like fuel particles during the sample fabrication process. Also, steel can demonstrate the applicability of X-ray CT technique for observing dispersion plates because iron attenuates photons in the X-ray energy range more than Al, like uranium [11]. Thus Type 304L stainless steel was selected as a surrogate for uranium fuel and the powder was prepared by gas atomization.

The stainless steel - aluminum dispersion plates were fabricated by the picture-frame method. First, atomized steel powder was mixed with fine Al powder. The mixing ratio was set to be the same as that of 9.0gU/cm^3 grade target. Two types of powder were used to prepare compacts for the CT optimization (45~150 micron), and for the analysis (300~500 micron). Compacts were fabricated to 15mm × 15mm × 2mm using a hydraulic press. Pressure was applied for 10 seconds at 400MPa. Compacts showed densification up to 95%. The compacts were then assembled with Al6061 cladding and then welded. The cladding consists of two cover plates and one frame. The size of each component was 45mm × 45mm × 2mm, and the frame has a blank space in the center to allow compact to be placed in. Welding was done by diffusion bonding using spark plasma sintering (SPS). The assembly was heated to 350ºC within 10 minutes, and then heated for a further 10 minutes. The heating process was performed in vacuum, with a pressure of 5MPa.

After welding, the sample was rolled using differential speed rolling machine (Daito Seisakusho/DBR 250 RDG). The rolling speed was set to keep the strain rate as 0.1s^{-1}. Samples were preheated in a furnace at 500ºC for 30 min, and reheated for 10 min between every passes. The rolling reduction ratio, defined as a ratio of thickness reduction during the rolling process, was set as Table I. Three samples underwent 9, 4, 3 times of hot rolling passes respectively, followed by cold rolling to a final thickness of 1.27mm.

Table I: Rolling condition

		A	B	C
Hot Rolling	#Passes	9	4	3
	Reduction	15%	30%	4-%
Cold Rolling		10%		

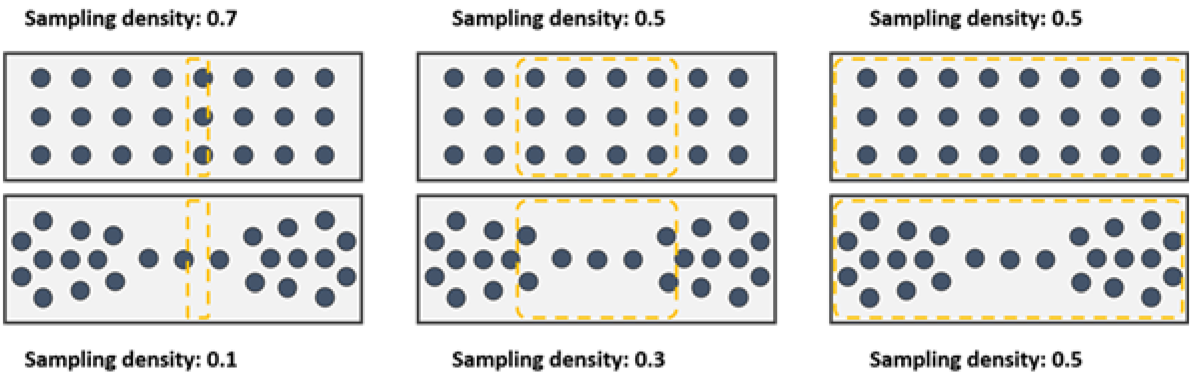

Figure 2. Basic concept of particle homogeneity estimation methodology

2.2 Data Acquisition

Data acquisition was done by X-ray CT observation. Power, X-ray energy, and resolution were the factors for selecting the CT equipment. X-ray source energy and resolution provides the contrast and distinguishability of particles on the data, while power and X-ray source energy provides the penetrating ability into sample to provide enough information for the analysis. Preliminary investigation was conducted to determine whether the CT technique is available for high-density LEU target observation. Three different CT equipment was tested and fabricated dispersion plates were observed using Nikon XT H 225 (Figure 2.), which was found to have high penetration and high contrast resolution due to its superb X-ray energy up to 225 kV.

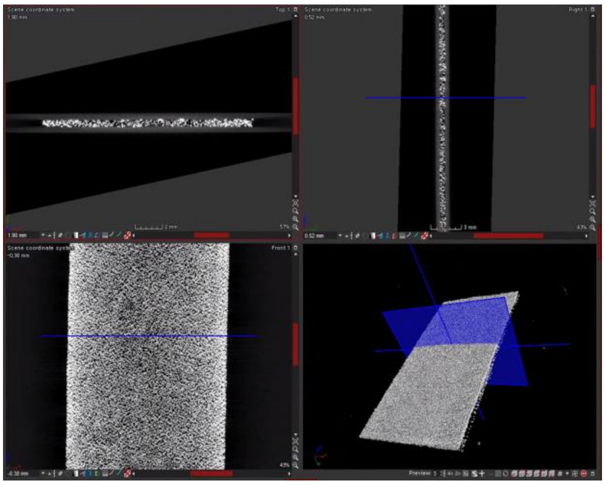

Figure 1. CT scan procedure by XT H 225

2.3 Particle Homogeneity Estimation

The basic principle for estimating the particle homogeneity is described in Figure 2. There are two images, a uniformly distributed image and an image with aggregation. As the yellow box size increases, the sampling density within the box converges to the particle fraction of the entire image. It should be noted that sampling density converges at a smaller cell size in case of uniformly distributed image, while it reaches at a larger cell size with aggregation. Based on this concept, the internal particle homogeneity can be obtained by estimating the sampling density according to the cell size increase. In this study, we obtained 3D data by CT scan results, thus the concept was expanded to 3D and the algorithm was developed to calculate the volume fraction.

The algorithm was constructed using MATLAB platform. Prior to the calculation, steel particles and Al matrix were binary-segmented. The volume fraction of steel particles in the segmented data was then calculated. After creating a cell in the 3D data stack, the volume fraction within the cell was calculated. Calculation was repeated with arbitrarily changing the position of the cell, and this procedure was repeatedly performed with increasing the cell size. Converging graph is obtained by plotting the calculated volume fraction results as cell size increases. Plotting standard deviation value of the calculated volume fraction for each cell size supports the convergence analysis. By plotting the different results, it is possible to identify the convergence difference and thus to determine the relative homogeneity difference.

3. Results

It was confirmed that X-ray sufficiently penetrates the fabricated dispersion plates thus the data contains enough information for the particle distribution analysis. And also steel particles were distinguishable on the CT data.

After the binary-segmentation, volume fraction calculation was performed. Data size was 500 × 20 × 600 pixels and for cell size from 5 pixels to 505 pixels by 10 pixels step, volume fraction of steel particles was calculated 1000 times for each cell size.

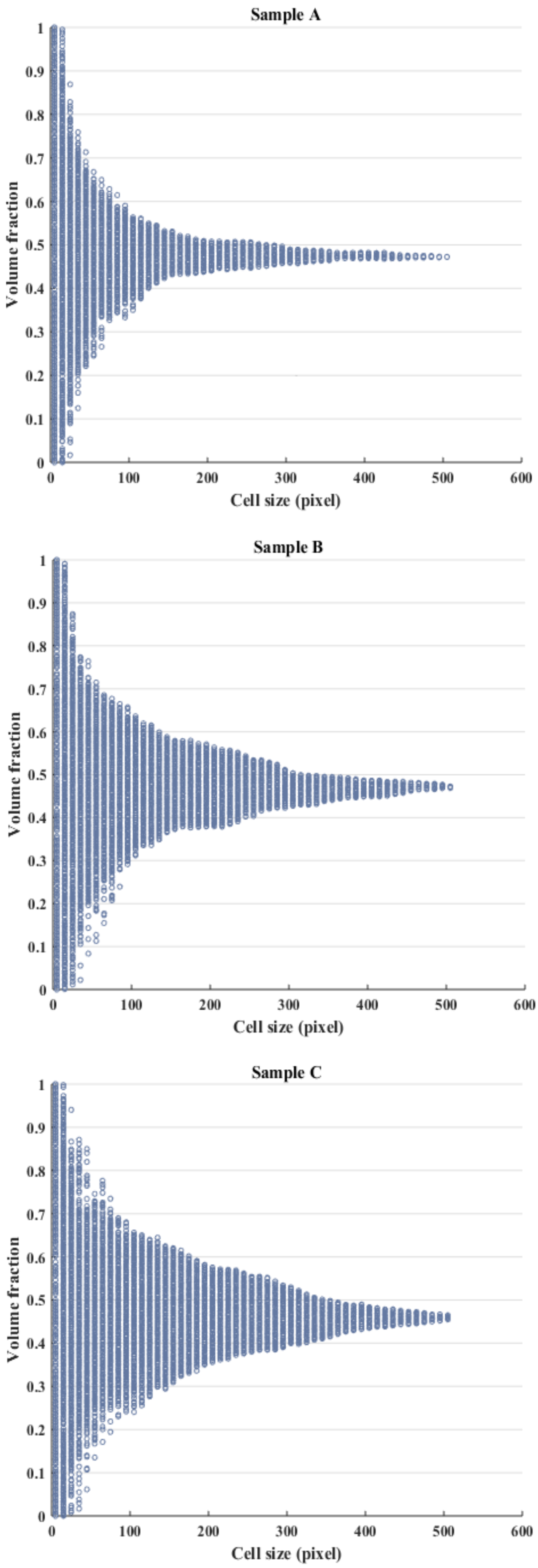

Figure 3. Particle volume fraction plot of sample A, B, C

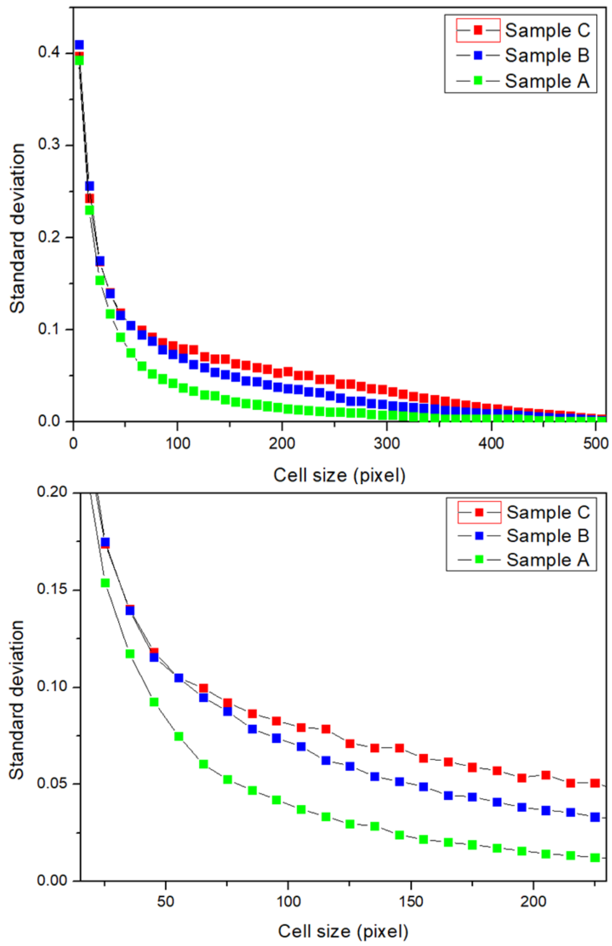

Figure 4. Standard deviation plot of fabricated samples

Result graphs (Figure 3.) showed the converging tendency depending on the rolling reduction ratio per pass. The volume fraction converges most rapidly in the sample fabricated with the lowest reduction ratio. It is more clearly shown in the standard deviation plot (Figure 4.) In this analysis, it was confirmed that the internal particle aggregation can be reduced by increasing the number of rolling passes.

During the rolling process, the sample enters roll gap with a certain contact angle, so the internal particles are pressed against the rolling direction. (Figure 5.) Since the stainless steel has a higher yield strength compared to aluminum, steel particles deform the matrix and migrate against the rolling direction, which causes the aggregation. The rolling reduction ratio affects both the magnitude, and the rolling direction component of the stress vector. Therefore under the same final reduction rate, by decreasing the reduction ratio per pass by increasing the number of rolling passes, the internal particle homogeneity was improved.

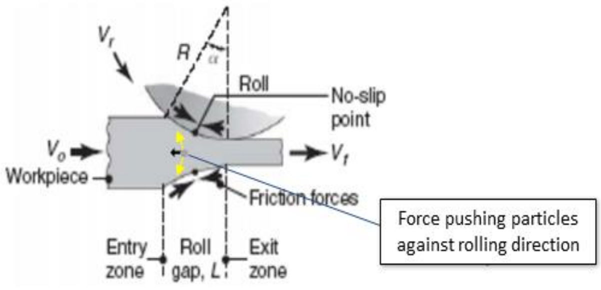

Fig. 5. Stress action in rolling process [12]

In summary, a 3D X-CT homogeneity analysis was done on the dispersion plates prepared with coarse size powder (300~500 micron) which is much larger than the conventional fuel particle size. Analysis results imply that increasing the number of rolling passes will enhance the internal particle homogeneity during fabrication process of high density target with coarse particles.

3. Concluding Remark

In this study, the applicability of X-ray CT technique to observe dispersion plates was investigated for the development of high density LEU target manufacturing technology. Stainless steel and aluminum was clearly distinguishable on CT data. An algorithm estimating the particle homogeneity from the 3D CT data was constructed. Furthermore, the effect of the number of rolling passes was analyzed using the established methodology. Dispersion samples were fabricated with 3 different conditions: 9 rolling passes with 15% reduction ratio, 4 rolling passes with 30% reduction ratio, 3 rolling passes with 40% reduction ratio. It was confirmed that the internal particle homogeneity was improved by setting smaller rolling reduction ratio per pass.

This analysis is expected to contribute to the stability and performance improvement of the high density LEU target by understanding the effect of the reduction ratio per pass controlled by the number of rolling passes.

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