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Identification and Particle Size Determination of 238Pu-bearing Particles via Alpha Spectrometry, Autoradiography and Scanning Electron Microscopy

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Introduction: In the event of an inhalation intake of $^{238}PuO_2$, establishing the size range of dispersed particles is imperative to determine the extent to which such materials enter, remain, and/or leave an exposed individual. Here we present an approach combining alpha spectrometry, autoradiography and scanning electron microscopy (SEM) with energy dispersive spectroscopy (EDS) of contaminated samples to locate, identify and determine the size distribution of ^{238}Pu-containing particles.

Samples and Methods: Initial autoradiography was performed on a sample cut from a lab coat (PPE) contaminated during a June 8, 2020 inhalation exposure incident at Los Alamos National Laboratory (LANL) to determine regions of greatest activity. Two identified high activity regions were sub-sampled and cut from the initial PPE within a glove bag to prevent dispersal of Pu particles. The two sub-samples were transferred into quartz tubes and placed in a muffle furnace where the samples were heated to 525 °C for 24 hours. This temperature was chosen to remove the majority of the organic lab coat material while having minimal impact on the morphology of PuO_2 particulates previously calcined at a higher temperature. Once cool, the resulting ash was suspended in ~5 mL of methanol and transferred into a 22 mL Savillex container to serve as a stock solution of particles.

Six particle mounts were prepared for alpha spectrometry and autoradiography on 1" stainless steel disks, with ½" circular carbon tape placed at the center of the disks. Using a hot plate set at 75 °C to accelerate drying, small aliquots (50-100 µL) of the methanol-ash solution were incrementally pipetted onto the carbon tape until a total activity of < 2500 dpm (estimated by gross alpha counting) was achieved.

The six particle mounts were counted by alpha spectrometry using an Ortec Ensemble equipped with 300 mm^2 UltraAS detectors to quantitatively determine the total ^{238}Pu activity on each mount. Navigational workflow for the remaining process is demonstrated in Figure 1. Starting with an autoradiograph collected using a GE Typhoon system (Figure 1a), to determine the spatial distribution of activity and select regions to investigate by SEM imaging (e.g. Figure 1b), a relatively low magnification setting was used to scan over the targeted area, in order to locate particles with a high Z signature (e.g. Figure 1b). Once a particle of interest was identified, a higher magnification was employed for imaging (e.g. Figure 1c), and EDS, which was used to determine if the particle was Pu-bearing (Figure 1d).

After radiography and prior to SEM, the samples were carbon coated to minimize charging during SEM analysis and to further bind the ashed PPE to the mount. All SEM imaging and EDS was done using a FEI Quanta 200F field emission SEM coupled with an EDAX Octane silicon drift detector. All EDS data were processed using EDAX TEAM software. Backscatter electron imaging (BSE) using an accelerating voltage of 15 kV and a spot size of 4 (spot size optimized for both imaging and EDS) was used to locate the particles, as high atomic number (high Z) species, such as Pu, appear brighter than the surrounding low Z ash material. Contrast and brightness of the SEM image were further adjusted to highlight the Pu particles from other high Z material.

Results and Discussion: A total of 11 Pu-bearing particles were identified from the six mounts. Particles ranged from 0.38 µm to 6.2 µm in size, with dimensions given in Table 1.

The autoradiography images of two samples where Pu-bearing particles were located via SEM (S1 and S2) were further analyzed to establish a correlation between the damage volumes in the radiography film and the size of each particle. For this process, each particle of significant activity was identified in the radiograph and numbered. The damage volume for each of these particles shown on the film was then calculated using GE's proprietary image analysis software. The damage volume of each particle was normalized, and the distribution was used to calculate the ^{238}Pu present in each particle as a fraction of the total ^{238}Pu activity measured by alpha spectrometry. The individual particle activity was converted back to a particle size using the theoretical density of $^{238}PuO_2$ (11.3 g/cm3) and under the assumption that all particles are spherical, are in the form of PuO_2 and are included in the analysis.

To validate this approach, particle sizes estimated from the radiography data were compared to the particle sizes measured by SEM (Table 2). Three confirmed $^{238}PuO_2$ particles were found in sample S1 by SEM, and two in sample S2. Although the number of comparable particles located by SEM imaging is limited, the size comparison is favorable.

The particle size distribution calculated for all 68 particles analyzed shows an average particle size of 0.20 – 0.30 µm, tailing up to 2 µm (Figure 2). The shape of this distribution approximates a log-normal distribution, which is typical of PuO_2 particle samples. The particles found by SEM tend to be at the larger end of this distribution. This discrepancy could result from larger particles being easier to find by SEM, or that the smaller particles in the radiography images are other materials containing a small activity of ^{238}Pu or other alpha emitting radionuclide. This is an area for further investigation, but it is reasonable to assume from the combined SEM and radiography results that most of the $^{238}PuO_2$ particles are < 2 µm.

Conclusions: The combination of autoradiogaphy, SEM, and EDS provides a novel approach for locating sparse Pu-bearing particles from contaminated materials (exposure events or otherwise). This allows for determining the size range of contaminant particles, which provides important constraints regarding how such materials may remain or pass through human bodies in the event of an inhalation exposure.

Table 1: Pu particle sizes from SEM imaging

Sample ID	Particle ID	Dimension 1	Dimension 2
S1	P1	0.98 µm	0.56 µm
	P2	2.67 µm	1.60 µm
	P3	1.20 µm	0.56 µm
S2	P1	6.16 µm	1.90 µm
	P2	0.96 µm	0.73 µm
S3	P1	2.02 µm	1.15 µm
S4	P1	1.07 µm	0.76 µm
	P2	0.77 µm	0.38 µm
	P3	0.78 µm	0.48 µm
S5	P1	2.08 µm	1.68 µm
S6	P1	0.79 µm	0.59 µm

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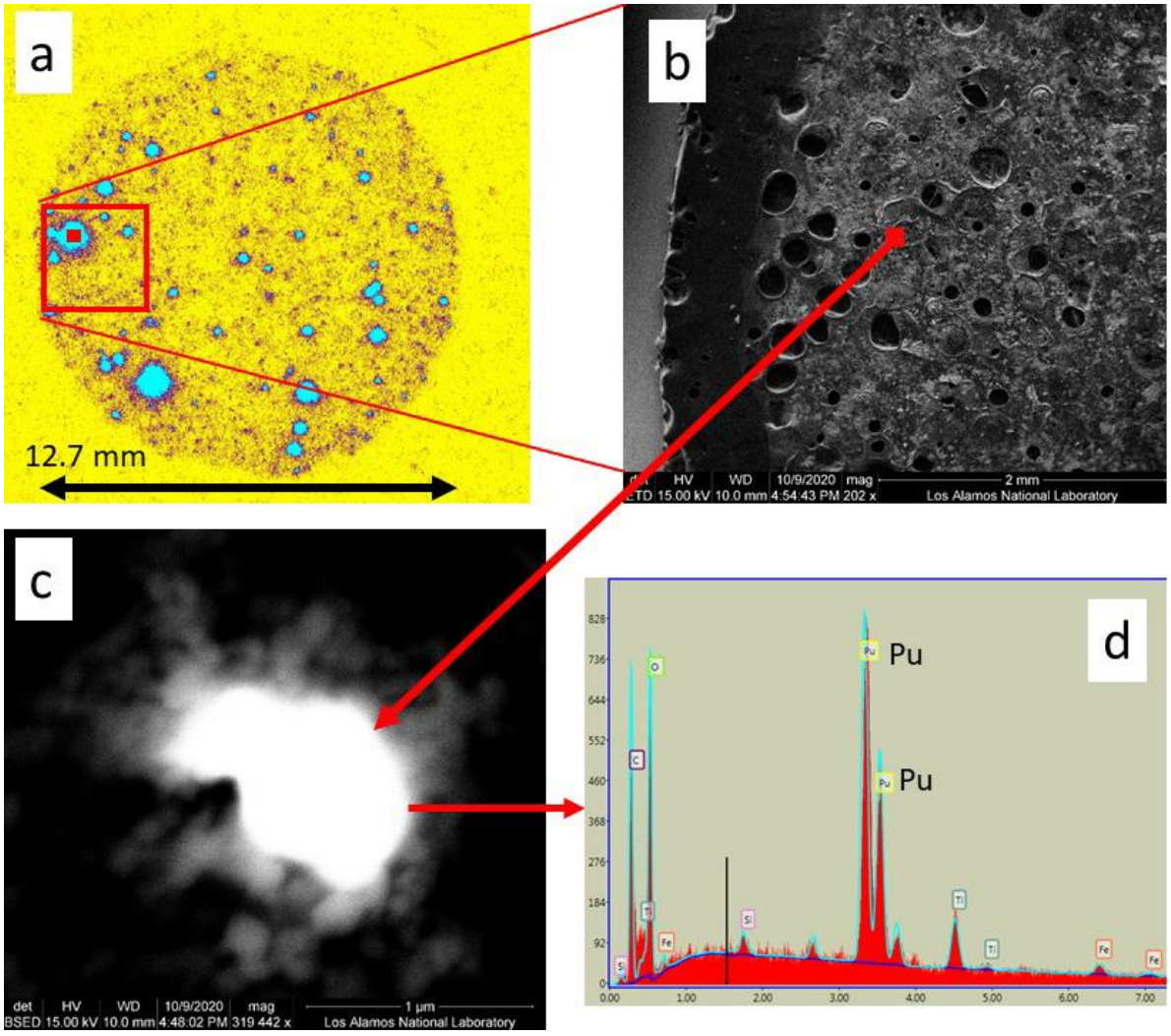

Figure 1. Example workflow for locating Pu-bearing particles. Starting with an autoradiograph image (a), regions of high activity are targeted and located within the SEM (b). Once high Z particles are located in BSE imaging (c), EDS is performed to generate a spectrum and determine whether or not the particle contains Pu (d).

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Table 2: Comparison of particle sizes determined by SEM and modeled by radiography

	Estimated Particle Size (μm)	
Particle	SEM (length x width)	Radiography (diameter)
S1-P1	0.98 x 0.56	0.71
S1-P2	2.67 x 1.60	2.1
S1-P3	1.20 x .560	0.73
S2-P1	6.16 x 1.90	1.2
S2-P2	0.96 x 0.73	0.69

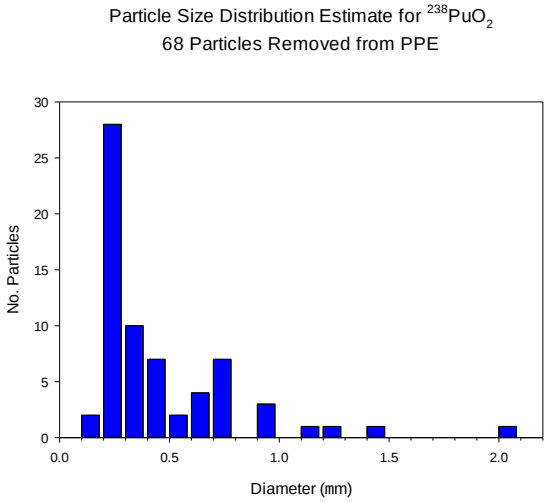

Figure 2. Particle size distribution estimated from radiography data and total ^{238}Pu activity on samples S1 and S2.

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