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Evaluation of Properties for Microsample Identification

November 2025

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Abstract

A study was conducted to determine if individual particle characteristics could be used to identify particles of interest, sub-samples, from bulk post-detonation debris. Three archived post-detonation debris samples were used for this effort. Particles from these samples were identified as active (produced fission tracks), and inactive (did not produce fission tracks), as the first defining characteristic. Morphology was the secondary characteristic to select particles for further study, i.e. spherical/non-spherical. Once particles were identified and isolated, they were characterized by optical microscopy for size in μm, number of fission tracks, morphology, transmitted light color, and reflected light color. Particles were then analyzed by scanning electron microscopy for morphology, elemental content, and compound identification. Raman spectroscopy was attempted on five particles with indeterminate results due to environmental mixing (heterogeneity) during the events of particle formation. Once all non-destructive analyses were completed all particles were analyzed by thermal ionization mass spectrometry to determine isotopic atom percents of plutonium and uranium, and an estimate of atoms of plutonium and uranium in each particle. An estimate of the ratio of uranium to plutonium was also obtained (U/Pu). Data analytics of the data from the particles showed that combining characteristics of the particles have a high probability of identifying particles of interest from bulk post-detonation debris samples.

Please note that this version of the report is an abridged version of the full report (Wagnon et al. 2025) that has been edited to be appropriate for public release.

Summary

Samples

Three archived post-detonation samples were selected for this study. They are designated as sample 256 (83 particles analyzed), sample 258 (85 particles analyzed), and sample 259 (74 particles analyzed). These samples are the particulate from the ashed portion of filter samples that were suspended in flexible collodion for isolation and characterization. The particulate suspensions were then put through the fission track method to identify particles that contained fissile/fissionable materials.

Particle Analysis

Using fission track stars in the Lexan detectors, particles were selected for further analysis, these particles are referred to as active particles, i.e. produce fission tracks. We also isolated and analyzed inactive particles, i.e. did not produce fission tracks, that had similar morphologies to the active particles. The isolated particles were then characterized using optical microscopy (OM). The characteristics determined by OM were, number of fission tracks, particle size (μm), transmitted light color, reflected light color, and morphology. Images of the fission track star (for active particles) and the isolated particle were obtained at OM. The isolated particles were then analyzed by scanning electron microscopy (SEM). Secondary electron and backscatter electron images were obtained for each analyzed particle. Energy dispersive spectroscopy (EDS) was used to determine the elemental content of each particle. An overscan technique was used to determine the elemental content of the main particle (parent) and, if appropriate, smaller areas were analyzed (daughter), i.e. different densities to parent in backscatter image. Once the non-destructive analyses were completed the particles were analyzed by thermal ionization mass spectrometry (TIMS). This analysis determined the uranium and plutonium isotopic content of the particles. Using the TIMS data an estimate of the number of atoms of uranium and plutonium as well as the elemental ratio (U/Pu) were determined.

Acknowledgments

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Acronyms and Abbreviations

EDS	Energy Dispersive Spectroscopy
EM	Electron Microscopy
NFTI	Nuclear Forensics Transformational Innovation
OM	Optical Microscopy
RPL	Radiological Processing Laboratory
SEM	Scanning Electron Microscopy
TIMS	Thermal Ionization Mass Spectrometry

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1.0 Introduction

The aim of this project was to analyze archived post-detonation debris material for physical attributes that may be unique to the material of interest found within the larger volume of sample material. These physical attributes may be exploited in subsequent separation techniques, to be identified in the future (see scoping study in section 2.0), to reduce the analysis sample size from grams to micrograms or less. If viable screening methods are identified, the majority of background material could be rapidly separated from residual materials of interest; this would not only reduce digestion volumes but also minimize background signal. Using these techniques could allow for shorter timelines from sample receipt to data reporting.

When the current standard methods for debris analysis were developed on samples collected during nuclear weapons testing, large samples (tens of grams) were required because of the detection limitations at the time and the dilute nature of weapons debris within the environmental samples. However, contemporary microanalysis techniques have shown promise in past work, and we contend that for several analytes, such methods should reduce the quantity of material required (i.e., milligrams to just micrograms following preconcentration). By using smaller samples and not requiring the dissolution of grams of refractory material, significant time savings can be achieved.

Bomb-rich micro-inclusions in debris samples may also offer the ability to reduce unwanted background in samples. The combination of new microanalysis techniques and reduced background could result in most required measurements being made very rapidly from microsamples that then can be used to answer priority forensics questions. Such questions are focused on the type, design, and origin of the item or device in support of attribution, enabling law enforcement to hold accountable those responsible. In addition, multiple bulk sample collections are currently required to make the fractionation (volatility) corrections needed for nuclear forensics assessments, which can quickly overwhelm laboratory capacities. Due to the heterogeneous nature of debris samples, we hypothesize that fewer collections would be required if we are able to create multiple enriched microsamples needed for fractionation corrections from a single bulk debris sample.

Using our mature fission track method for identifying actinide material, where fission products should be collocated, we isolated particles of interest in archived debris samples. These particles were examined by optical microscopy to identify physical characteristics that may be indicative of isotopes of interest for debris characterization, including their size and shape. Once the optical interrogations were complete, Raman spectroscopy was performed on select particles. Raman spectroscopy is a non-destructive analytical technique that can be used to identify the crystallographic structure of microparticles that may contain isotopes of interest. Particles were then examined via scanning electron microscopy for elemental content via energy dispersive spectroscopy. This elemental information can be used to identify any relationship between major-/minor- elemental composition and the possible presence of isotopes of interest for debris analysis. From the characteristics determined by these methods subsamples of the original bulk sample may be identified and used for more targeted isotopic analysis then can be achieved with the bulk sample.

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2.0 Methods

2.1 Fission Track Detection Method

In the fission track method, the particulate of interest is suspended in flexible collodion so that the particulate is well dispersed in the final suspension. This suspension is then plated onto Lexan slides and allowed to polymerize. The Lexan/collodion pairs are then bundled together and heat sealed in poly sheeting. These bundles are then neutron irradiated in a reactor. These irradiations were performed at Washington State University. Once the bundles return from the reactor the collodions are separated from the Lexans. The Lexans are then “developed” to show the fission track damage to the Lexan detector. The Lexan slides are then scanned by OM to determine the location of the fission track stars, and these locations are used to determine the location of the particle that contained the fissile/fissionable material in the collodion associated with that Lexan.

2.2 Optical Microscopy

Using the Lexan scanning information, i.e. fission track stars, and morphology, particles are selected for isolation and analysis. Selected particles are isolated from collodion using a scalpel blade. Cleaned particles are then characterized for size (µm), morphology, transmitted light color, and reflected light color. The particle is then transferred to a carbon planchet and sent on to electron microscopy for analysis. For our first round of analyses, we chose to isolate particles with spherical morphology and fission tracks, i.e. active particles. A spherical morphology indicates the particles were probably exposed to a high temperature environment. A follow-on effort was to isolate and analyze inactive spherical particles. To address an even wider array of particles, we decided to interrogate each sample to find an active to inactive ratio for the particles. For each sample a single collodion was chosen, and 10 random fields of view were scanned for active and inactive particles. The sum of active and inactive particles was used to determine a ratio of active to inactive particles for each of the samples. Using the determined ratios an additional 50 particles were isolated for each sample, i.e. for a ratio of active/inactive of 0.25 – 10 active particles and 40 inactive particles would be isolated for analysis.

2.3 Raman Spectroscopy

Five particles were selected for confocal Raman analyses at the Radiological Processing Laboratory (RPL) using a Rensihaw inVia system. For the pilot analysis, particles with diameters between 3.1 -14.3µm and 30 – 7,500 tracks were selected. Raman analyses were performed using multiple laser wavelengths (532, 758, 833 nm) to achieve an optimal spectrum. Despite using multiple wavelengths, all of the assessed particles exhibited significant fluorescence that impeded structural identification. The lack of diagnostic spectra and severe fluorescence could reflect a high-level of crystallographic disorder due to post-det conditions, however, attributing null analytical results to an underlying process is challenging and, if multiple laser wavelengths are required for each analysis, time consuming. Because a rapid screening technique is desired in this application, further Raman spectroscopy characterization was not pursued on this effort.

2.4 Electron Microscopy

For each isolated/cleaned particle the following analyses were performed using a scanning electron microscope. Secondary electron images were obtained, these images show the morphology of the particle at a finer scale than that obtained at OM. Backscatter electron

images were obtained, these images show variation in the Z number of elements present in the particle, i.e. areas with high uranium content will be brighter than areas with high silicon content. Using the backscatter electron images as a map the particles were analyzed for elemental content using energy dispersive spectroscopy (EDS). If the backscatter image was homogenous then an overscan analysis was performed and the elemental information is assigned to the parent particle id. If the backscatter image was heterogeneous then an overscan analysis was performed on the predominant density area of the particle and assigned the parent particle ID. For smaller areas with different densities, a finer EDS analysis was performed and each of the areas was assigned a daughter ID. These daughters are included and/or occluded materials associated with the parent/main particle. Using the EDS spectra, elemental percentages were assigned for each area analyzed. A stoichiometric analysis of the elemental percentages was performed to determine compounds present in the analysis regions.

2.5 Thermal Ionization Mass Spectrometry

Once the OM and EM analyses were completed the particles were removed from the carbon planchet and placed onto pre-carburized rhenium TIMS filaments for isotopic analysis. For each of the particles an isotopic analysis for plutonium and uranium was attempted, this included inactive particles, i.e. no fission tracks in the Lexan detector for the particle being analyzed. The TIMS analyses were performed in two runs, the first run is for plutonium, and the second run is for uranium. The data from these analyses were used to determine isotopic ratios, atom percents for each isotope and to estimate the elemental ratio (U/Pu) and estimate the number of atoms of each element present in the particles analyzed.

2.6 Data Analytics

The compiled spreadsheet containing results from OM, EM, and TIMS analysis was used to identify correlations among the techniques. Pairwise correlations were computed using the Pearson correlation coefficient. A machine learning model was used to examine the ability of OM information to predict particle activity. When training the model, 80% of the data were used for training and 20% were used for testing. When possible, the train-test sets were evenly split based on class labels (i.e., stratified sampling). Numerical inputs to the model were first scaled by the maximum absolute value in the corresponding feature set. Categorical features were one-hot encoded. The model was generated using the Decision Tree Classifier implemented in the Scikit-Learn Python package. A maximum tree depth of 5 was found to give the highest accuracy with minimum overfitting.

2.7 Scoping Study of Separation Techniques

A scoping study was conducted to evaluate other particle phenomena that may influence debris composition and particle behavior in the environment. The scoping study was focused on surface effects such as electrostatic interaction or self-charging from radioactive decay along with magnetic properties caused by incorporation of constituents like iron. These effects may have variable influence on particle behavior where environmental conditions may impact how the various phenomenon affect particle composition and size. More details can be found in the stand-alone report (currently in draft).

3.0 Results and Discussion

3.1 Particle Down Selection Using OM

We first set out to answer the question “Can OM be used to down select particles that may contain fissile/fissionable material?” For this, we used the transmitted light color, reflected light color, and morphology of isolated particles recorded by the microscopist. A particle was considered active if TIMS showed the presence of U-235.

Figure 1 shows pie charts of the recorded colors for active and inactive particles based on the sampling source. Source 256 has notably different optical characteristics than sources 258 and 259. Active particles from source 256 are largely composed of silicates (as determined by EM), which often produce green, yellow, or mottled transmitted light and colorless reflected light. Active particles from source 258 include fewer silicates and more iron-aluminum oxides. Corresponding, the transmitted green particles disappear, leaving silicates that produce mottled or yellow transmitted light. The iron-aluminum oxide particles tend to produce rust color transmitted light and metallic reflected light. Active particles from source 259 are mostly comprised of iron-aluminum oxides that produce rust color transmitted light and metallic reflected light. The remaining active particles are silicates. In all three samples, inactive particles were composed of carbon compounds, silicates, and metal oxides.

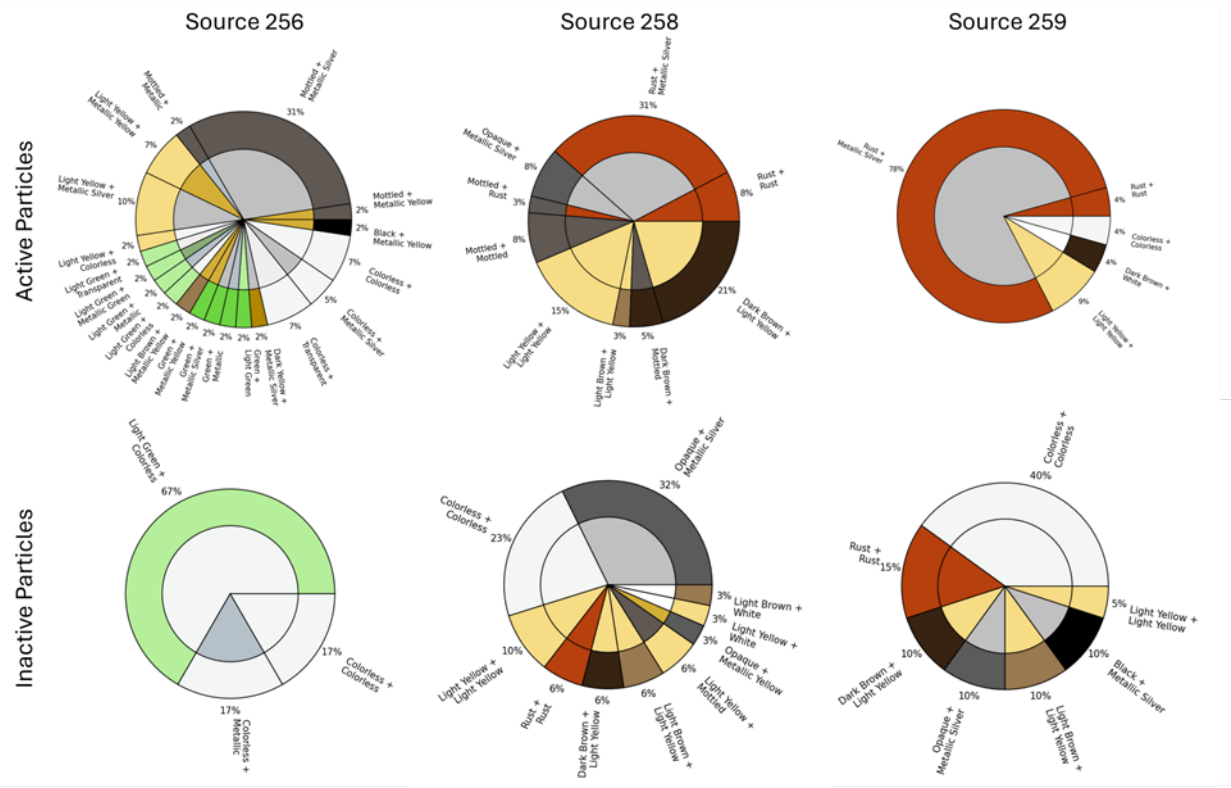

Figure 1 Recorded Transmitted Light Color (outer ring) + Reflected Light Color (inner ring) for Active (top row) and Inactive (bottom row) Particles Based on the Sampling Source (columns)

Due to the different characteristics of particles samples from the 3 source locations, we then examined correlations between transmitted and reflected light colors from OM and proportion of U measured by TIMS for active particles (Figure 2). Particles from source 256 showed the weakest correlations between U content and color, with slight positive correlations between yellow transmitted light and U content and metallic yellow reflected light and U content. Slight negative correlations were observed for light green transmitted light and U content and colorless reflected light and U content. Particles from source 258 showed slight positive correlations between U content and rust color, dark brown, and mottled transmitted light. Slight negative correlations with U content were observed for colorless reflected light, colorless transmitted light, and opaque transmitted light. Particles from source 259 showed stronger correlations between U content and rust color transmitted light and metallic silver reflected light, and negative correlations between U content and colorless transmitted and reflected light.

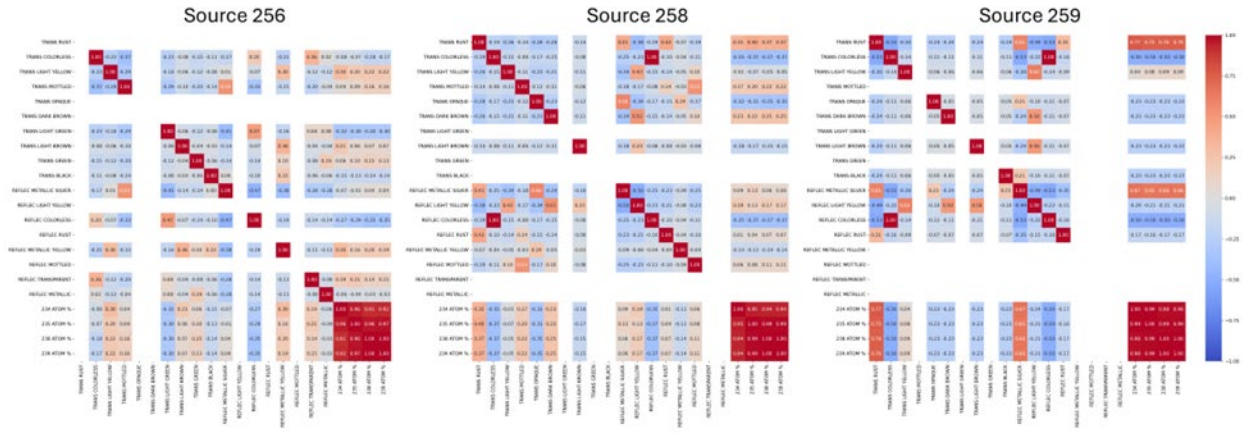

Figure 2 Correlation Matrices for Active Particles from Each Sampling Source

Considering the observed correlations, if down selecting particles based on OM, particles that produce colorless transmitted or reflected light should be avoided as these particles tend to be inactive (though some active particles may be missed depending on the sampling source). Particles showing rust color transmitted light and metallic yellow reflected light should be prioritized as these particles are more likely to show activity.

3.2 Predicting Activity from OM Data

A Decision Tree Classifier was used to predict if the particle was likely to show activity or not based on the transmitted and reflected light colors, particle morphology, particle size, and source ID. An average test set accuracy of 67% was obtained for three trials with different random seeds. The most important features for accurate activity predictions were particle size, when the source ID was 259, metallic silver reflected light, opaque transmitted light, transparent transmitted light, and rust color transmitted light. Therefore, these colors may be used to indicate positive or negative particle activity, while other colors do not provide predictive power.

A possible cause of the limited predictive power of OM color is the small size of the dataset, the variety of colors given, and the subjective color description. Notably, factors such as local geology and the distance between the sampling site and test site could also affect the apparent correlations between particle coloration and size, respectively, with activity.

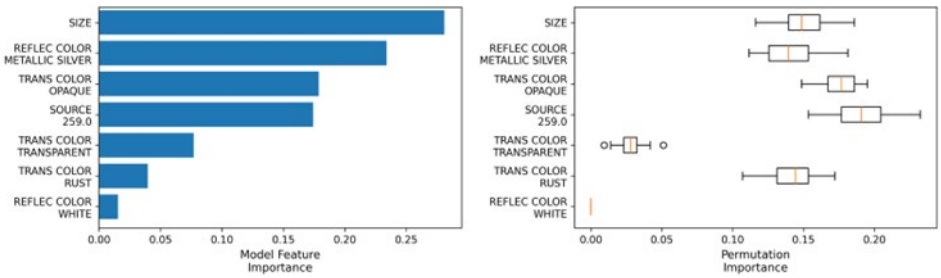

Figure 3 Importance of Features in the Decision Tree Classifier Trained to Predict Start Type

3.3 Predicting Star Type from OM Data

A Decision Tree Classifier was used to predict the Lexan star type based on the transmitted and reflected light colors, particle morphology, particle size, and source ID. An average test set accuracy of 73% was obtained for three trials with different random seeds. The most important features for accurate star type predictions were source ID and particle size (Figure 4). Rust-color transmitted light was the only color feature to have comparable importance in the model. Reflected metallic silver and light yellow and transmitted mottled and green showed minor importance. Although the test set accuracy was fairly high, OM data alone is not a good predictor of star type due to the primary importance of source ID and particle size in the classification.

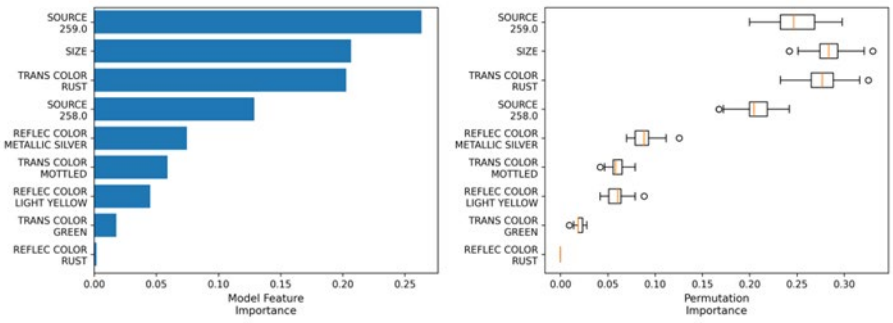

Figure 4 Importance of Features in the Decision Tree Classifier Trained to Predict Start Type

4.0 Conclusions

Using a combination of post-detonation particle characteristics, we believe there is a high probability there could be successful separation of material of interest from the environmental background of debris material from an event. An example would be the combination of spherical/semi-spherical morphology, rust color for transmitted light, and metallic yellow color for reflected light (see Data Analytics, section 2.6). If a large filter sample or representative synthetic filter sample was available, a combination of optical imaging and rapid activity measurements (e.g. autoradiography), could be used to rapidly screen thousands of active and inactive particles. Relative to the particles analyzed in this study – which represent a relatively small sample set that was tailored to spherical particles – using a combination of spatially correlated OM and autoradiography measurements could increase the size of the ML data set by several orders of magnitude and result in more accurate and less biased predictions.

5.0 References

Wagnon, Ken, et. al. 2025, Evaluation of Properties for Microsample Identification – Final Report FY2025, PNNL-38666

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