

LA-UR-17-26776

Approved for public release; distribution is unlimited.

Title: Characterization of Morphology using MAMA Software

Author(s): Gravelle, Julie

Intended for: Report

Issued: 2017-08-02

Disclaimer:

Los Alamos National Laboratory, an affirmative action/equal opportunity employer, is operated by the Los Alamos National Security, LLC for the National Nuclear Security Administration of the U.S. Department of Energy under contract DE-AC52-06NA25396. By approving this article, the publisher recognizes that the U.S. Government retains nonexclusive, royalty-free license to publish or reproduce the published form of this contribution, or to allow others to do so, for U.S. Government purposes. Los Alamos National Laboratory requests that the publisher identify this article as work performed under the auspices of the U.S. Department of Energy. Los Alamos National Laboratory strongly supports academic freedom and a researcher's right to publish; as an institution, however, the Laboratory does not endorse the viewpoint of a publication or guarantee its technical correctness.

Characterization of Morphology using MAMA Software

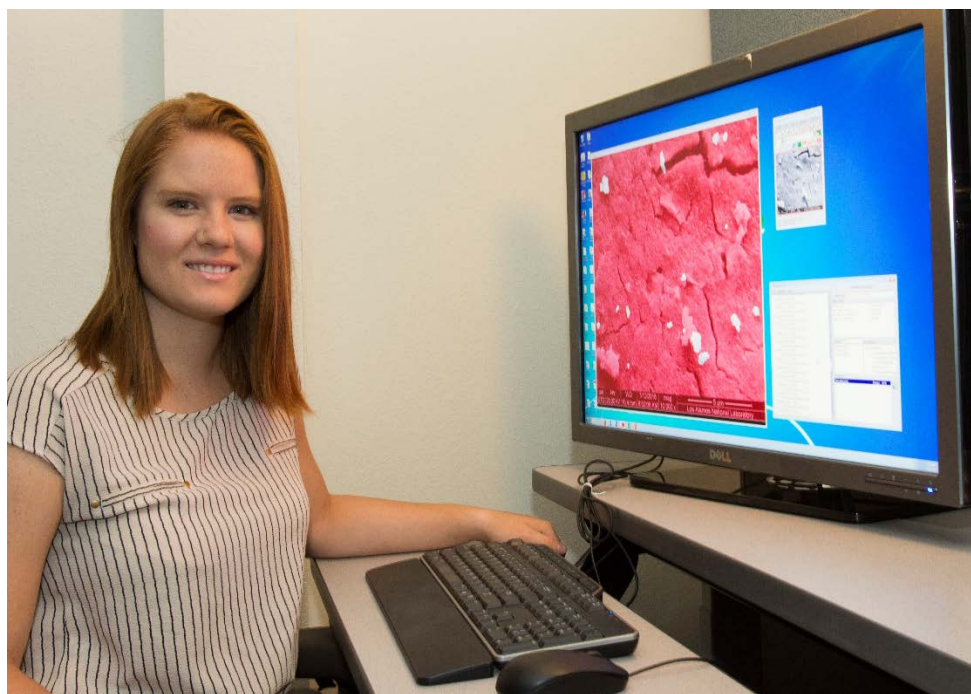
Julie Gravelle

Hosting Site: Los Alamos National Laboratory

Mentor: Marianne P. Wilkerson

Requires LA-UR

The MAMA (Morphological Analysis for Material Attribution) software was developed at the Los Alamos National Laboratory funded through the National Technical Nuclear Forensics Center in the Department of Homeland Security. The software allows images to be analysed and quantified. The largest project I worked on was to quantify images of plutonium oxides and ammonium diuranates prepared by the group with the software and provide analyses on the particles of each sample. Images were quantified through MAMA, with a color analysis, a lexicon description and powder x-ray diffraction. Through this we were able to visually see a difference between some of the syntheses. An additional project was to revise the manual for MAMA to help streamline training and provide useful tips to users to more quickly become acclimated to using the software. The third project investigated expanding the scope of MAMA and finding a statistically relevant baseline for the particulates through the analysis of maps in the software and using known measurements to compare the error associated with the software.



Using the MAMA software for image analysis.

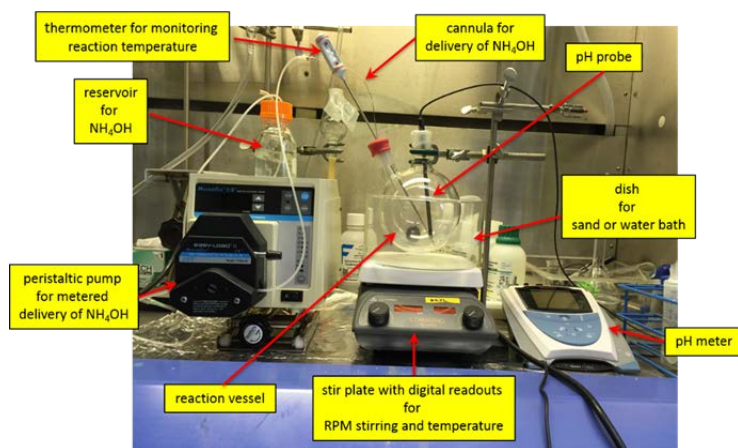
1. Internship Project

During this internship, I worked on several different projects dealing with the MAMA software. The revision of the user manual for the MAMA software was the first project I was able to work and collaborate on. I first learned how to use the software by getting instruction from a skilled user at the laboratory, Dan Schwartz, and by using the existing user manual and examples. After becoming accustomed to the program, I started to go over the manual to correct and change items that were not as useful or descriptive as they could have been. I also added in tips that I learned as I explored the software. The updated manual was also worked on by several others who have been developing the program. The goal of these revisions was to ensure the most concise and simple directions to the software were available to future users. By incorporating tricks and shortcuts that I discovered and picked up from watching other users into the user guide, I believe that anyone who utilizes the software will be able to quickly understand the best way to analyze their image and use the tools the program offers to achieve useful results.

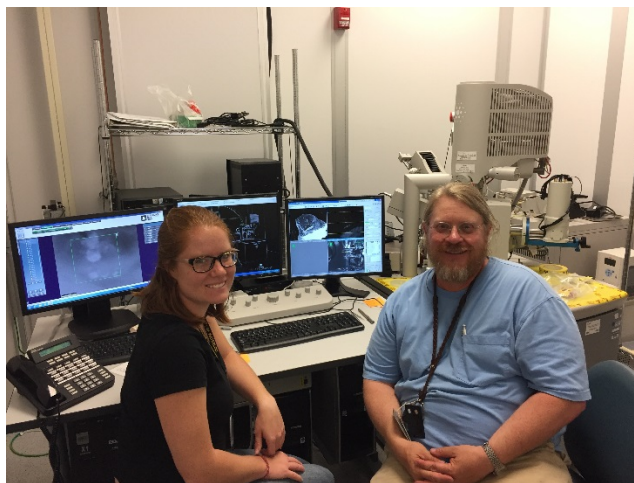
Next, I was tasked with quantifying images of ammonium diuranates for projects that other employees had begun work on in the lab. This was a central part of the experiment because the conclusions drawn were based around the analyses carried out using the software. I presented this project in a poster format at the first Interagency Technical Nuclear Forensics Program Review in Oak Ridge, Tennessee during the week of July 24, 2017.

In order to analyze the images, the ammonium diuranates first had to be synthesized under different conditions. Eighteen different variations of temperature, stir rate, ending pH, flow rate, and concentration of uranium were generated. Each variation was synthesized using the set up below to monitor temperature, flowrate, and stir rate. The different variables were chosen from the table below.

[U]:[NO ₃] (mg/mL)	Stir rate (rpm)	Flowrate (mL/min)	Ending pH	Temperature (°C)
50	170	2.5	5	21.5
100	280	5	8	35
200	400	7.5	11	50



Ending pH was used to determine reaction completion. The dry samples were then imaged and analyzed. I was able to go to the NISC (Nonproliferation and International Security Center), under escort, and learn how scanning electron microscopy (SEM) images are taken and see the process of ensuring quality images are taken (shown below). Analysis was carried out in four forms: color, morphology, lexicon, and powder x-ray diffraction.



To perform the color analysis of the samples, I used a Munsell Soil Color Charts (2009 Revision). The dried samples were displayed and compared to the guide. I recorded the specific value, hue, chroma and corresponding color.

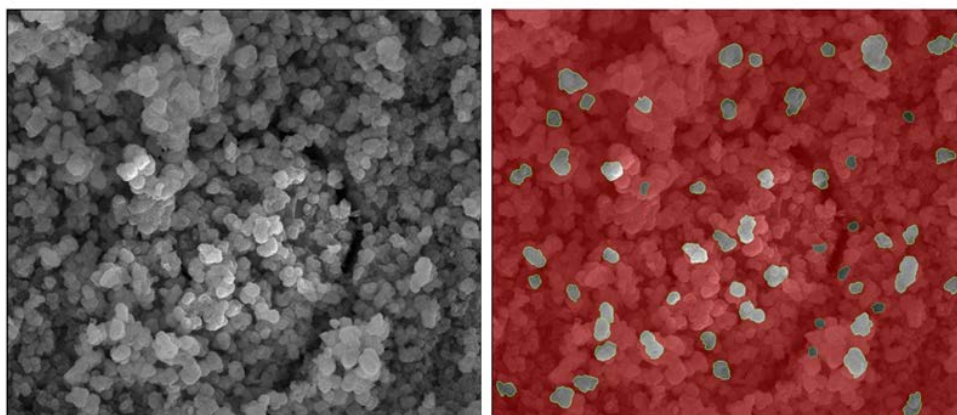
In terms of color, all but three of the samples were categorized as 'yellow'. Within the broad category of yellow, however, the hues of the samples ranged from 2.5 Yellow-Red to 5 Yellow. The remaining three

samples were determined to be shades of red or yellow with one 'red' (9), one 'reddish yellow' (13), and one 'yellowish red' (5).



The Munsell Charts allow each of the colors to be identified in terms of the general color (hue), the amount of lightness (value), and the strength of the color (chroma).

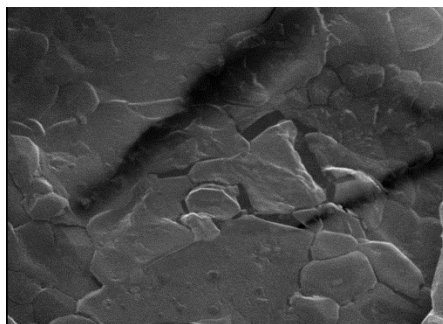
I completed morphological analysis with the MAMA software. SEM images were taken with an FEI Quanta 200F Field Emission Scanning Electron Microscope of each sample at various locations on the sample with different magnifications. These images were uploaded to MAMA, where particles selected using a flowchart were segmented from the background and quantified. The mean values of each result were recorded.



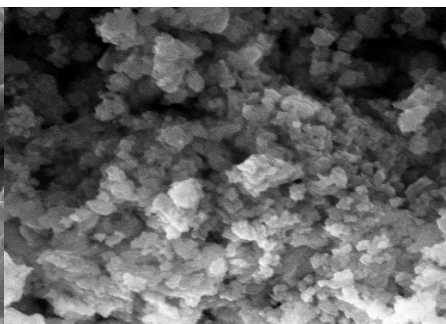
An example of an image of ammonium diuranate before and after segmentation in MAMA.

The lexicon analysis was also completed in the MAMA software using the built-in lexicon descriptors. Each of the set of images for a sample in a given area were described by going through the lexicon and choosing the best fit from each section. They were applied to all the magnifications of that particular set and updated for a particular image if a lower magnification allowed the identification of a different morphological feature. Most samples were classified as 'Massive/Clumped', 'Agglomerate', and 'Rounded or Blocky'.

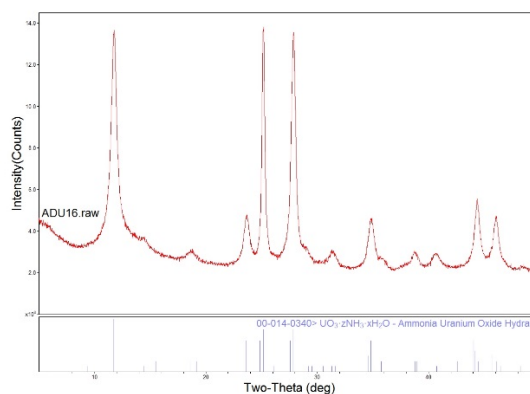
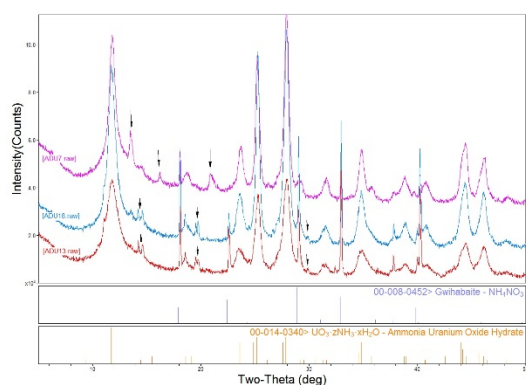
This sample was identified as 'Flattened', 'Platy', 'Melted' and 'Cracked/Fractured'.



This sample was identified as 'Somewhat Smooth' with 'Rounded/Subrounded' particles.



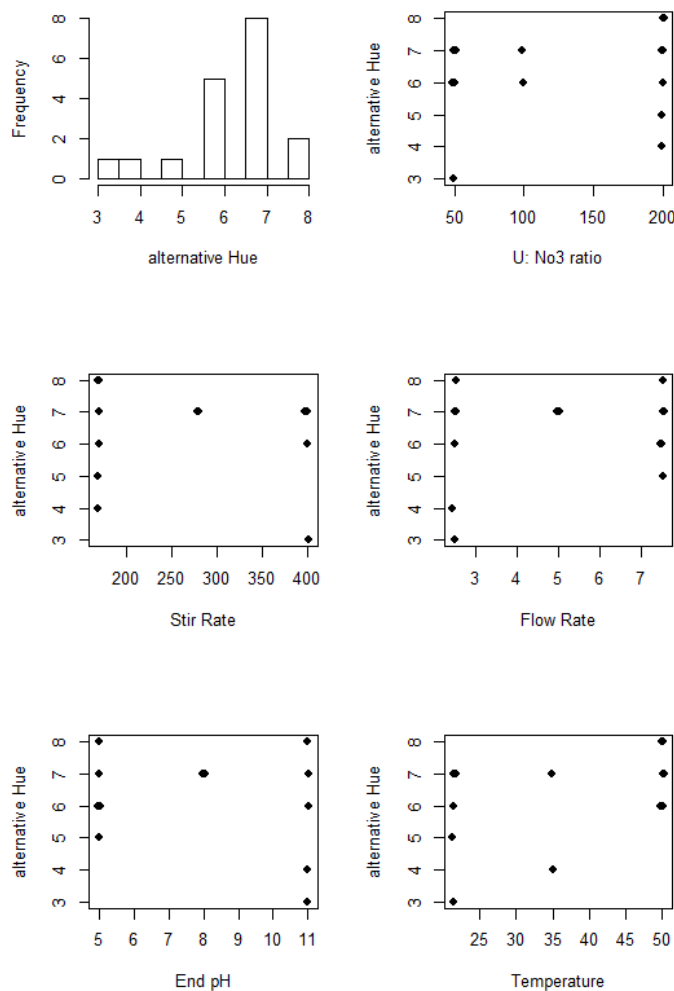
The p-XRD patterns were taken and analyzed by an analyst at the lab. Samples 7, 13, and 18 (left) contained $\text{UO}_3 \cdot z\text{NH}_3 \cdot x\text{H}_2\text{O}$, ammonium nitrate, and some unknown peaks. Sample 16 (right) contained only $\text{UO}_3 \cdot z\text{NH}_3 \cdot x\text{H}_2\text{O}$. All other samples contained $\text{UO}_3 \cdot z\text{NH}_3 \cdot x\text{H}_2\text{O}$ with some ammonium nitrate present. Further analysis is needed to determine the numerical values of z and x .



I tabulated the data from all four analyses, and it will be studied in the future for statistically significant correlations. My initial findings showed that the samples with a stronger red hue were formed in a lower temperature at a higher pH, while the samples with the stronger yellow hue were formed at the highest temperature with a high concentration of uranium dissolved in the nitric acid. Preliminary analysis of the lexicon descriptors showed that samples made at lower temperatures resulted in a mixed product that contained 'Rounded/blocky' particles as well as 'Crystalline/faceted' particles.

The tabulated data was given to a statistician who worked to create models and graphs to attempt to discover relationships between the variables and the data produced. Initial statistical analysis of the data showed that the models have some predictive power, but none can really explain all of the patterns from

the data. Significantly more data would be necessary to draw a more definite conclusion. An example of the models produced follows.



Larger value for hue is seen for lower ending pH and for higher temperature.

In addition to creating a poster to present this work, I also began work on a scientific paper that will be completed following more thorough statistical analyses. I drafted the experimental sections about how each of the analyses were performed and the results and conclusions sections to discuss the findings of the research.

I also used MAMA to analyze images of plutonium oxide particles that were taken for a different project. Using the same technique, the particles were segmented and quantified. The data was then tabulated, and before and after images, along with the data, were used in a scientific talk at the program review.

The next project I worked on was the implementation of MAMA software on maps. Maps provide a reliable background for analysis because the segments (around countries, landmarks, and natural features) are already known. The area, length, angles, and exact borders have already been calculated. Because of this, it is useful to apply the segmentation and quantification of the MAMA software to maps. By using maps as the images to be quantified, the results can then be statistically compared to the known values for those boundaries. Through this statistical comparison, an error can now be associated with the software and extrapolated to the future analyses performed on nuclear samples. To begin, I found images of maps of various places, such as the United States, Canada, Europe, and Africa, making sure they had lines marking the outlines of states, provinces and countries. I then segmented the maps and tried to ensure the segments followed the borders as closely as possible. Issues that arose in this project dealt with the quality of images used to segment. It was difficult to know if the borders I was segmenting were the correct ones due to over-pixelated images and broad lines that left out detail in the true borders of the country. Also, the scale of the images was difficult to overcome in the software because the ruler has a preset pixel to micron scale, but the images were showing thousands of kilometers of distance. The software, due to this, did not have extremely accurate results due to rounding of the larger numbers than it normally deals with.

2. Impact of Internship on My Career

The summer internship has shaped my future very strongly. I find that this summer revealed new possibilities in the field that are not widely publicized because most people, when thinking about forensics, assume the only careers are in law enforcement and do not think of the field as requiring research in methods and in applications.

In terms of my academics, I plan to use the knowledge and skills I have learned at the Los Alamos National Laboratory to further my understanding in the field of forensics and continue to learn about the instrumentation, methods, and application of new forensic research in the context of law enforcement. I plan to enroll in a graduate program that will allow me to apply the research I have done, and likely will still do.

Though I do not continue to plan a career solely in research, the internship was a useful experience and allowed me to develop an appreciation for and understanding of the ground breaking work within and outside the realm of nuclear forensics research.

The most interesting event I attended while working at the laboratory was the National Technical Nuclear Forensics Center program review. I was able to attend four days of talks about research being done all over the country in different kinds of labs and learn about what is going on in the field of nuclear forensics. In addition, I presented a poster at the unclassified poster session which allowed me to learn about how to disseminate scientific works as well as see others' posters about other research in the field.

The program review was an impactful experience because of the sheer scope of research and projects that I was exposed to during my time in Oak Ridge. Talks about different materials, methods, and instruments were presented during the review from scientists working at national labs and several universities. Having so much information presented to me about nuclear forensics helped me broaden my knowledge about the options that exist in forensics and what sort of techniques are being practiced around the country.

The Los Alamos National Laboratory hosted several events to benefit all employees, including frequent lectures and laboratory-wide events that allowed several different organizations within the laboratory to communicate their information. One specific event that I found beneficial were the Nuclear Lunches that took place weekly on site. The summer seminar allowed for open discussion about safeguards, nonproliferation, and chemistry as it related to the topic chosen for that week. Some of the topics included: IAEA Inspection Measurements; Uranium Mining, Milling, and Conversion; and Reprocessing. Each week, an expert in the topic would lead the discussion, but people from all backgrounds (scientists, students, post-doctoral employees) gathered to bring their own ideas and thoughts on the subject for that week. These lunches were an opportunity to learn about and discuss the other aspects of nuclear research, outside the scope of just forensics or chemistry.

Another lecture I attended was one by Doug Berning, a program manager for forensics projects at the laboratory. He spoke on post-detonation forensics, which was a new side of the research for me, as my

projects focused on pre-detonation forensics. The lecture included an overview of the Department of Energy Forensics Operations Team and discussions on nuclear detonation scenarios.

Some of the other opportunities I took advantage of while in Los Alamos include a talk by the director of the lab to the students about how we fit in and grow at the laboratory. I also attended a lecture about the history of nuclear weapons and the evolution of the stockpile to modern day non-proliferation efforts. I learned about the history of plutonium-238 and its production and peaceful applications. By learning more about the laboratory, the materials I worked with, and the history of their use,

If this research were to be continued under the scope of the Department of Homeland Security, it would prove useful in many aspects of criminal investigation involving nuclear proliferation. The morphological analysis of particles of nuclear origin is a useful investment of time and expertise because it provides insight into the origin of the material and the conditions of its creation and storage. The future of ammonium diuranate project will involve statistical analysis of the data collected from all four analyses to determine if there is any significant difference based on the different variables used in the syntheses. If research is to be continued in this field, it is possible that a library of materials and conditions for each material could be developed to ease identification of the material and its origin. If illegal substances are then recovered, it would be possible to discover what the substance is, where it was illegally removed from, and possibly what conditions or locations it was stored in prior to recovery.

3. Acknowledgments

I would like to express gratitude to my mentor and group members who provided me with invaluable knowledge and support this summer. I would also like to thank those who collaborated with me in using the MAMA software. Finally, I would like to express my deepest appreciation to the Los Alamos National Laboratory for providing the resources necessary to complete this research, as well as the DNDO for their student summer program. This research was supported in part by an appointment to the U.S. Department of Homeland Security (DHS) Domestic Nuclear Detection Office (DNDO) Summer Internship Program. This program is administered by the Oak Ridge Institute for Science and Education (ORISE) through an interagency agreement between the U.S. Department of Energy (DOE) and DNDO. ORISE is managed by ORAU under DOE contract number DE-SC0014664.

4. References

1. Ainscough, J. B.; Oldfield, B. W. "Effect of ammonium diuranate precipitation conditions on the characteristics and sintering behavior of uranium dioxide" J. Appl. Chem. 1962, 12, 418-424.
2. Doi, H.; Ito, T. "Significance of physical state of starting precipitate in growth of uranium dioxide particles" J. Nucl. Mater. 1964, 11(1), 94-106.
3. Woolfrey, J. L. "The Preparation and calcination of ammonium uranates – A literature survey" September 1968, Australian Atomic Energy Commission
4. Tomazic, B.; Samarzija, M.; Branica, M. "Precipitation and hydrolysis of uranium (VI) in aqueous solutions – VI investigation on the precipitation of ammonium uranates" J. Inorg. Nucl. Chem. 1969, 31, 1771-1782.
5. Janov, J.; Alredson, P. G.; Vilkaitis, V. K. "The influence of precipitation conditions on the properties of ammonium diuranate and uranium dioxide powders" J. Nucl. Mater. 1972, 44, 161-174.
6. Steeper, T. J.; Zink, J. C. "Particle-size distribution of ammonium diuranate precipitate" Proc. Okla. Acad. Sci. 1974, 54, 83-87.
7. Woolfrey, J. L. "The Preparation of UO₂ Powder: Effect of Ammonium Uranate Properties" J. Nucl. Mater. 1978, 74, 123-131.
8. Urbanek, V.; Sara, V.; Moravec, J. "Study of formation and composition of ammonium uranates" J. Inorg. Nucl. Chem. 1979, 41, 537-540.
9. Rajagopal, S.; Asari, T. P. S.; Iyer, C. S. P. "Particle size analysis of ammonium uranate prepared by conventional and homogeneous methods of precipitation and their corresponding oxides" J. Nucl. Mater. 1996, 227, 300-303.
10. Narasimha Murty, B.; Balakrishna, P.; Yadav, R. B.; Ganguly, C. "Influence of temperature of precipitation on agglomeration and other powder characteristics of ammonium diuranate" Powder Technology 2001, 115, 167-183.
11. Manna, S.; Roy, S. B.; Joshi, J. B. "Study of crystallization and morphology of ammonium diuranate and uranium oxide" J. Nucl. Mater. 2012, 424, 94-100.
12. Hoyt, R. C. "Precipitation kinetics of a continuous precipitator, with application to the precipitation of ammonium polyuranate" 1978 Iowa State University, Ph.D. Thesis.

Olsen, A. M.; Richards, B.; Schwerdt, I.; Heffernan, S.; Lusk, R.; Smith, B.; Jurrus, E.; Ruggiero, C.; McDonald, L. M. "Quantifying α -Morphological Features of α -U₃O₈ with Image Analysis for Nuclear Forensics" *Anal. Chem.* 2017, 89, 3177-3183.

13. Tamasi, A. M.; Cash, L. J.; Eley, C. J.; Wilkerson, M. P. "A lexicon for consistent description of material images for nuclear forensics" *J Radioanal Nucl Chem.* 2016, 307, 1611-1619.