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Contractor For U.S. Department of Energy, Office of River Protection, under Contract DE-AC27-08RV14800

**Release Stamp**

DATE:
Jan 30, 2018
HANFORD
RELEASE

**Clearance Review Restriction Type**: public

1. **Doc No:** RPP-RPT-60542 **Rev.** 00

2. **Title:**
Final Report for X-ray Diffraction Sample Preparation Method Development

| | |
|---|---|
| 3. **Project Number:** ☒ N/A | 4. **Design Verification Required:**<br>☐ Yes ☒ No |
| 5. **USQ Number:** ☒ N/A<br>RPP-27195 | 6. **PrHA Number** **Rev.** ☒ N/A |

7. **Approvals**

| **Title** | **Name** | **Signature** | **Date** |
|---|---|---|---|
| Clearance Review | RAYMER, JULIA R | *RAYMER, JULIA R* | 01/30/2018 |
| Document Control Approval | PORTER, MARY | *PORTER, MARY* | 01/30/2018 |
| Originator | ELY, TOM M | *ELY, TOM M* | 01/15/2018 |
| Other Approver | KRAFT, DOUGLAS P | *KRAFT, DOUGLAS P* | 01/11/2018 |
| Responsible Manager | COOKE, GARY A | *COOKE, GARY A* | 01/15/2018 |

8. **Description of Change and Justification**

Initial Release.

9. **TBDs or Holds** ☒ N/A

10. **Related Structures, Systems, and Components**

| a. **Related Building/Facilities** ☒ N/A | b. **Related Systems** ☒ N/A | c. **Related Equipment ID Nos. (EIN)** ☒ N/A |
|---|---|---|
| | | |

11. **Impacted Documents – Engineering** ☒ N/A

| **Document Number** | **Rev.** | **Title** |
|---|---|---|
| | | |

12. **Impacted Documents (Outside SPF):**

N/A

13. **Related Documents** ☒ N/A

| **Document Number** | **Rev.** | **Title** |
|---|---|---|
| | | |

14. **Distribution**

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Title: Final Report for X-ray Diffraction Sample Preparation Method Development

Information Category:
☐ Abstract ☐ Journal Article ☐ Summary
☐ Internet ☐ Visual Aid ☐ Software
☐ Full Paper ☐ Report ☑ Other SPF Report

Publish to OSTI? ☑ Yes ☐ No

Trademark/Copyright "Right to Use" Information or Permission Documentation: Yes ☐ NA ☑

Document Number: RPP-RPT-60542, Rev. 00 Revision 00

Date: ~~December 2017~~ JRR 01/30/2018

Author: Ely, Tom M

## Part II: External/Public Presentation Information

Conference Name:

Sponsoring Organization(s): WRPS

Date of Conference:

Conference Location:

Will Material be Handed Out? ☐ Yes ☑ No

Will Information be Published? ☐ Yes ☑ No *(If Yes, attach copy of Conference format instructions/guidance.)*

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      <comments>Reviewer's Comments Date: 01/12/2018 12:46 PM Step Name:
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        60542, rev 00 reviewed 1/12/2018 and can be publicly released without
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        RPP-RPT-60542 Final Report for X-ray Diffraction Sample Preparation
        Method Development Performer: O'Hagan, Michael (h6070759) No
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**RPP-RPT-60542 , Rev. 00**

# Final Report for X-ray Diffraction Sample Preparation Method Development

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U.S. Department of Energy Contract DE-AC27-08RV14800

| | | | |
|---|---|---|---|
| EDT/ECN: | DRCF | UC: | N/A |
| Cost Center: | 2LH00 | Charge Code: | N/A |
| B&R Code: | N/A | Total Pages: | 69 |

Key Words: Sample Preparation, Method Development, Saltcake, PLM, SEM, XRD, ALARA

Abstract: WRPS-1500790, "X-ray Diffraction Saltcake Sample Preparation Method Development Plan/Procedure," was originally prepared with the intent of improving the specimen preparation methodology used to generate saltcake specimens suitable for XRD-based solid phase characterization. At the time that this test plan document was originally developed, packed powder in cavity supports with collodion binder was the established XRD specimen preparation method. An alternate specimen preparation method less vulnerable if not completely invulnerable to preferred orientation effects was desired as a replacement for the method.

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DATE:
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# Final Report for X-ray Diffraction Sample Preparation Method Development

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Date Published
December 2017

Prepared for the U.S. Department of Energy
Office of River Protection
Contract No. DE-AC27-08RV14800

## EXECUTIVE SUMMARY

This report captures the detail of the work performed to develop a suitable X-ray diffraction specimen preparation method that is less susceptible to preferred orientation effects than the packed powder cavity method that has been the established method used at the 222-S Laboratory. In addition to this, the report also captures the body of precursor and associated specimen preparation efforts that enabled the specimen preparation method development work to be performed.

In the early sections of the report, precursor topics related to the selection of the instrument, configuration, and measurement settings, as well as the subsequent calibration process, are mentioned. These topics, in addition to familiarity with the theoretical operating principles of X-ray diffraction (i.e., review of X-ray diffraction reference text books), were considered to be critical foundational elements that had to be established prior to the onset of specimen preparation activities. Among the critical ideas described in text references was the specimen preparation method selected for further development using saltcake phases.

The instruments used in the course of this study are briefly described as well, and data interpretation concepts are presented.

Over the two-plus year span of this project, elements of the test plan evolved as did the tools used to prepare specimens. These details are described within the report, including the benefit to the project. Data quality improvements were the primary objective of the study; however, many “As Low As Reasonably Achievable” (ALARA) benefits were also integrated into the development of tools and the evolution specimen preparation methodology.

Considerable effort was put into the documentation of tool design and construction. Component materials (i.e., description needed to re-order parts) and their role in tool design and/or specimen preparation are described in sufficient detail for a knowledgeable scientist to be able to reproduce the associated construction or preparation processes.

The results of the various experiments performed revealed that the method was successful in terms of addressing the original problem (i.e., minimizing preferred orientation effects in the diffraction primarily introduced by the specimen preparation process used). At the same time, vulnerabilities were revealed. In the first rounds of testing, analysis of the diffraction data seemed to indicate a degree of analytical robustness that appeared to be impervious to accidental disruptions of the specimen deposit after the deposition process was initially completed. When they occurred, these disruptions were typically encountered during the mask removal stage of the specimen preparation process. However, in the last round of testing, which used a five-phase mixture, diffraction results in conjunction with analysis of the microscope images of the deposited powders revealed that phases with low angle content in the multi-phase mixture frequently were not detected when mechanical defects in the deposit were observed. While this vulnerability is significant and should not be ignored, the specimen preparation process developed allows for the rapid generation of a replacement specimen in routine analytical situations.

One interesting side effect was noted in the course of this project. The deposited powders transformed from an adhered particulate form to a contiguous "crust" form presumably by interacting with the adherent layer. This interaction/transformation process did not have a detectable adverse effect on the diffraction data, but it did unambiguously change the observable deposit surface morphology. The nature of this interaction is not understood completely and was only briefly investigated during this study. This unintended side effect did have the effect of reducing the potential for spreading the deposited powder inadvertently after the transformation process was completed. This will be beneficial in the preparation of all tank waste specimens in the future as the completely bound (transformed crust) deposit clearly has better ALARA characteristics in terms of spreadability of contamination.

Although the work activities within the scope of the original test plan have been completed, future work is anticipated. In this work, the scope of the specimen preparation process could be expanded to include other established solid phase characterization methods such as scanning electron microscopy and polarized light microscopy. At the same time, the established vulnerability of the current method could also be addressed to further enhance the robustness of this specimen preparation process.

With regard to ALARA elements developed as part of this project, many of the tools developed would have been much less evolved were it not for the extensive collaboration that occurred throughout the 222-S Laboratory complex. The results of many of the more significant tools and their associated ALARA benefits are also described in this document.

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## Acronyms and Abbreviations

| | |
|---|---|
| ALARA | As Low As Reasonably Achievable |
| LED | light-emitting diode |
| MDI | Materials Data Incorporated |
| PDF 4+ | Powder Diffraction File, version 4+ |
| PLM | polarized light microscopy |
| PVC | polyvinyl chloride |
| XRD | X-ray diffraction |

## Units

| | |
|---|---|
| cm | centimeter |
| ºF | degrees Fahrenheit |
| mL | milliliter |
| mm | millimeter |

# 1. BACKGROUND

The work described in this final report and the original test plan (WRPS-1500790, X-ray Diffraction Saltcake Sample Preparation Method Development Plan/Procedure) represent one component of a larger general effort to improve X-ray diffraction (XRD)-based identification of crystalline phases common to tank waste at Hanford. A more complete representation of the topics and issues that influence the generation and interpretation of XRD-based crystalline phase identification is shown in Figure 1. X-ray Diffraction Procedures: For Polycrystalline and Amorphous Materials, (Klug and Alexander 1966) and Elements of X-ray Diffraction, (Cullity 1978) are well known textbook references that address XRD operating principles as well as provide discussions relating to data processing and analysis methods. Specimen preparation methods are also addressed in these textbooks, but in more general terms than the work effort described in this report.

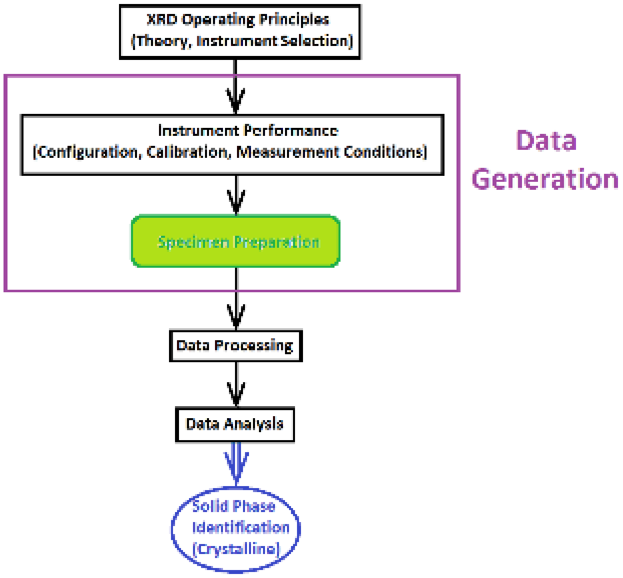

**Figure 1. This diagram indicates the complete context of considerations that impact the successful identification of crystalline phases. Specimen preparation is highlighted in green to emphasize the primary topic of this study.**

WRPS-1604673, "Evaluation of New X-ray Diffraction Instrument Systems," addresses many of the pertinent details associated with the selection of an XRD instrument within the context of routine work performed at the 222-S Laboratory at Hanford.

WRPS-1503787, "Calibration of MiniFlex II"; WRPS-1504345, "Calibration State of MiniFlex II"; WRPS-1600741, "Instrument Calibration Methods for the MiniFlex II"; ATS-LT-507-105, Rev A-0, "XRD Instrument Calibration for the Rigaku MiniFlex II"; RPP-PLAN-60909 Rev.00B, "MiniFlex II Calibration Template"; and WRPS-1703206, "Silver Behenate Powder and Silver Membrane Filters," all represent work performed at the 222-S Laboratory relating to the definition and monitoring of proper instrument performance characteristics.

A Practical Guide for the Preparation of Specimens for X-ray Fluorescence and X-ray Diffraction Analysis (Burke et al., 1998), generally describes (in section 4.5.7 "Alternative Flat Surface Techniques, Top Dusted Mounts," p. 149) the specimen preparation technique that was modified and optimized for saltcake specimens in the method development study described herein. Many of the major results of this study were presented in TOC-PRES-17-2845-VA, "Method Development for X-ray Diffraction Analysis of Thin Powder Deposit Samples of Simulated Radioactive Tank Waste," which was presented in August 2017 at the Denver X-ray Conference in Big Sky, Montana.

Efforts to further develop and optimize data processing and analysis methods will continue to be considered in the future. However, the instrument calibration and specimen preparation efforts that precede data acquisition frequently generate data of sufficient quality so that advanced analytical software such as Materials Data Incorporated (MDI) Jade 9.7[1] yields reliable phase identification without invoking advanced data processing methods.

The specimen preparation method development work described in this report was performed systematically but also within a larger context of related work that includes the topics represented in Figure 1. While these precursor topics will not be explicitly discussed in this report, they formed the data quality foundation that enabled the specimen preparation method development activities to be carried out successfully.

## 2. INTRODUCTION

WRPS-1500790, "X-ray Diffraction Saltcake Sample Preparation Method Development Plan/Procedure," was originally prepared with the intent of improving the specimen preparation methodology used to generate saltcake specimens suitable for XRD-based solid phase characterization. At the time that this test plan document was originally developed, packed powder in cavity supports with collodion binder was the established XRD specimen preparation method used at the 222-S Laboratory. While this specimen preparation is one of many specimen preparation techniques in common usage in analytical laboratory environments and is also described in the textbooks cited in the previous section, the packed powder specimen preparation method is also widely known to be susceptible to generating preferred orientation artifacts that interfere with the identification of solid crystalline phases. Correspondingly, an alternate specimen preparation method less vulnerable if not completely invulnerable to preferred orientation effects was desired as a replacement for the method in use at the time. Both saltcake and sludge phases are common among solid tank waste materials here at Hanford. However, saltcake phases were selected for this method development study as they, as a class of waste materials, are more vulnerable to preferred orientation effects.

The six saltcake phases originally selected for evaluation: (1) were commonly found in tank waste saltcake samples in the past and (2) had one or more crystal habit(s) that was vulnerable to preferred orientation-related distortion of XRD data. Sodium phosphate dodecahydrate was eliminated in the course of the study (see section 4, Exceptions to the Test Plan). Examples of

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[1] Jade 9.7 is a product of Materials Data Incorporated, Livermore, California.

typical particle morphologies for the remaining five saltcake phases are shown in Figure 2 (component images in Figure 2 were excerpted from LAB-RPT-15-00005 R0, *Hanford Tank Waste Particle Atlas*).

Correspondingly, the main goal of the test plan was to develop a specimen preparation method that yields reproducible high-quality XRD data on simulated and eventually on actual saltcake samples, while controlling the spread of contamination and maintaining or improving as low as reasonably achievable (ALARA) practices. Work performed during the course of this project (spanning May 2015 through May 2017) was recorded in HNF-N-859-1, "Research Projects," (e.g., simulant synthesis and microsieve construction) and also in HNF-N-710-1, "Rigaku MiniFlex II[2] XRD" (e.g., XRD measurements).

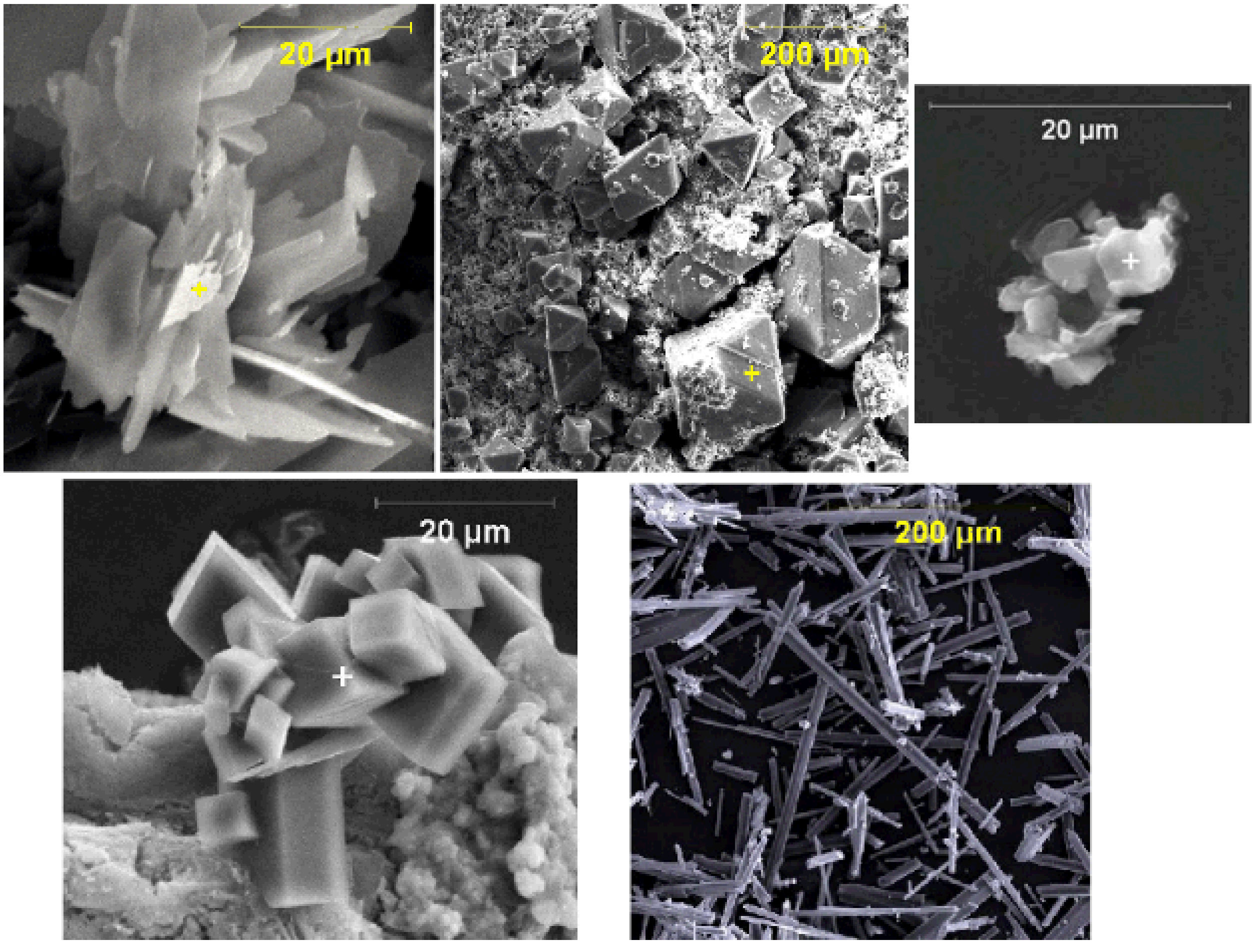

**Figure 2. Non-equant saltcake phase particle morphologies. Top-Left: Thermonatrite; Top-Middle: Natrophosphate; Top-Right: Burkeite; Bottom-Left: Nitratine; Bottom-Right: Natroxalate.**

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[2] MiniFlex II is a product of Rigaku Americas Corporation, The Woodlands, Texas.

# 3. SPECIMEN PREPARATION METHOD AND INSTRUMENTS SELECTED FOR DEVELOPMENT

A general outline of the specimen preparation method further developed during this project is presented in section 4.5.7, "Alternative Flat Surface Techniques, Top Dusted Mounts" (Burke et. al. 1998). Paraphrasing from that text …the use of this specimen preparation method is advised when preferred orientation effects result from other preparation methods or when small amounts of sample are used to prepare specimens. In the case of actual tank waste analyte materials, both of those conditions are routinely satisfied. The text also advises the use of petroleum jelly as the adherent grease.

The major refinements to the textbook method evaluated (1) the use of a removable mask in conjunction with a single crystal substrate to control the geometry (i.e., area and adherent layer thickness) of the deposit and (2) replacing a 100-mesh sieve with both a modified sieve and an alternate powder distribution method that did not involve a sieve.

## 3.1 Diffraction Measurements

### 3.1.1 Measurement Conditions

With the exception of the reference data shown in Figure 3, all of the diffraction data presented in this report were collected using identical measurement conditions on a Rigaku MiniFlex II with a 6-position sample stage and graphite monochromator using copper K-alpha radiation and the specimens were spun during data acquisition. However, the measurement conditions shown in Figure 3 are only different in terms of the scan speed. For this method development study, a speed of 1.0 degree per minute was used to acquire all diffraction data. Typical tank waste characterization measurements performed at the 222-S Laboratory are performed at a scan speed of 0.1 degree per minute (i.e., 10x slower) with all other measurement conditions as indicated in Figure 3 and also as described in ATS-LT-507-103, "222-S Laboratory X-Ray Diffractometry (XRD) Using The Rigaku MiniFlex II." As this study involved measurements on more than 100 specimens, the scan speed was increased to 1.0 degree per minute to accommodate the available instrument time.

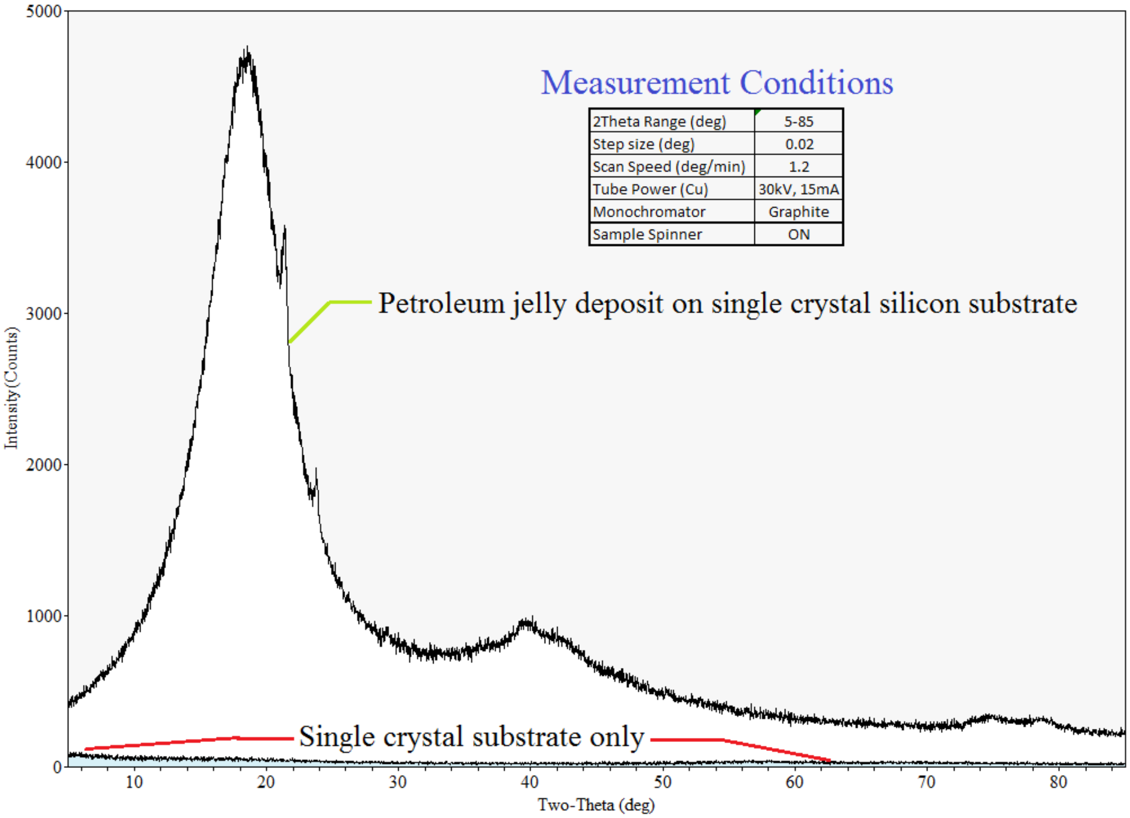

**Figure 3. Reference data sets are shown for a typical single crystal substrate and a similar single crystal silicon substrate coated with petroleum jelly.**

### 3.1.2 Interpretation of Diffraction Data

Clearly from the data shown in Figure 3 (and by design), the single crystal substrate does not generate a strong diffraction signature. However, for the sake of proper interpretation, the two reference patterns cannot be directly subtracted. The miniscule substrate scattering response is non-linearly attenuated (i.e., exponential absorption of X-ray coupled with a geometrically varying beam path during the measurement process) when the petroleum jelly over-layer is present.

Similarly when powder is deposited on the petroleum jelly, the powder layer will reduce the diffraction signature of the petroleum jelly adherent layer and further diminish the substrate contribution of the observed diffraction data to insignificance.

## 3.2 Optical Microscopy Instruments

### 3.2.1 Polarized Light Microscopy

Polarized light microscopy (PLM) methods were used exclusively to verify that the three synthesized saltcake simulants were phase pure prior to the extraction of precipitated solids from partially evaporated chemical solutions. An Olympus BX51 TRF polarized light microscope was operated in accord with procedural instructions described in ATS-LT-519-107, “222-S

Laboratory Polarized Light Microscopy," and information relating to the images acquired was recorded in HNF-N-464-1, "Polarized Light Microscope Instrument Log 1." Interpretations during live review of simulant precipitates in solution, and subsequently of the images generated, were provided by a trained operator (M. E. LaMothe).

### 3.2.2 Optical Microscopy

Low magnification microscope images of prepared specimens were acquired using an Olympus SZX-16 microscope using external light-emitting diode (LED) lights for illumination, a 0.5X objective lens, and 0.8X zoom lens settings. The images were typically acquired immediately after the specimen mounts were prepared and then the specimen mounts were installed in the MiniFlex II for XRD measurements. In some cases, the diffraction measurements were performed first and microscopy was performed subsequently. Image files were routinely copied from the microscope control computer and saved in the diffractometer control computer in the same data folder that included the associated diffraction data. Both data sets were recorded in HNF-N-710-1.

## 4. EXCEPTIONS TO THE TEST PLAN

### 4.1 Saltcake Simulant Phases

The test plan document was originally released in March 2015 and subsequently revised two times (e.g., Waste Stream Fact Sheets were updated). Synthesis of the saltcake simulants began in May of 2015. Over the same period of time, informal observations were made during the course of an unrelated study that revealed one of the simulant salt materials (sodium phosphate dodecahydrate) was subject to dehydration transformation (see HNF-N-859-1, page 26, dated 7/9/2015). As a result, a collaborative decision was made to eliminate this material from the study.

Of the five remaining saltcake simulants, three were synthesized (natrophosphate, burkeite, and thermonatrite) as commercially produced reagents were not available. All three synthesized simulant phases were evaluated using PLM prior to extracting precipitated solids from the solution each time it was processed to extract precipitate solids. Here PLM evaluation was used as a means of ensuring that the solids being produced were phase pure. However, the thermonatrite saltcake simulant yielded 140% of the expected product yield and was later (post extraction processing of the solids) independently determined not to be phase pure, based upon analysis of the corresponding XRD data. Representative PLM images of the successfully synthesized simulants are shown in Figure 4.

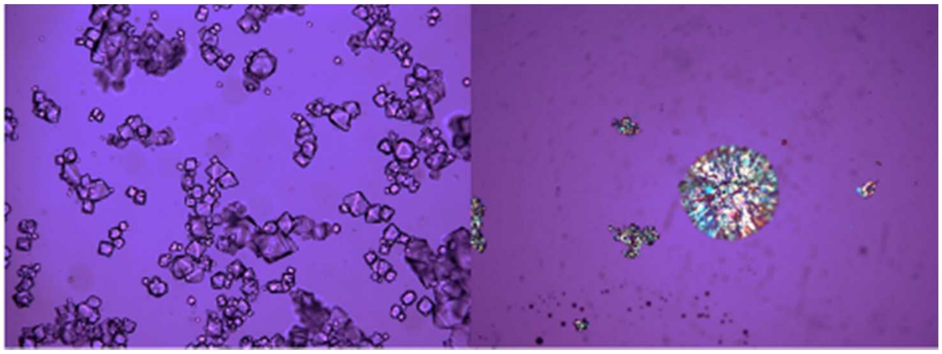

**Figure 4. Example photos of natrophosphate (left) and burkeite (right) synthesis solutions preceding recovery of precipitated solids.**

In the case of the synthesized thermonatrite saltcake simulant, trona and nitratine were also identified by XRD (see Figure AII-9, Appendix II, for phase identification data). During synthesis, the original recipe included in the WRPS-1500790 plan/procedure was modified (sodium carbonate monohydrate was replaced by anhydrous sodium carbonate, and the recipe was scaled up to produce more product simulant). Correspondingly, it is likely that a mistake was made during the preparation of the simulant solution resulting in the unexpected and undesired secondary phases.

Both thermonatrite and trona have very similar crystal habits and optical properties (see Figure 5) (*the right caption of Figure 5 was excerpted from LAB-RPT-15-00005)*. Meanwhile, it is likely that the nitratine identified by diffraction precipitated during the vacuum filtering process used to separate the saltcake solids from the liquid solution. Consequently, the presence of this phase was not evident in the PLM images acquired during the synthesis process.

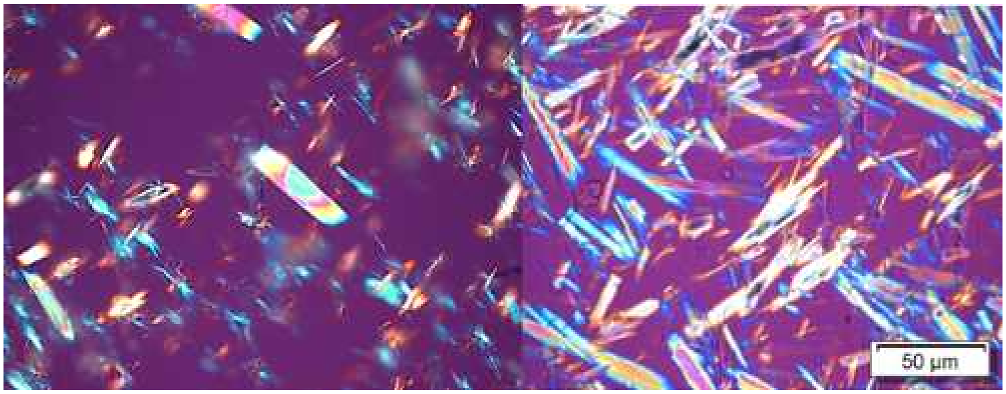

**Figure 5. Thermonatrite in solution (left) and trona (right) are shown in this figure.**

### 4.2 Staff Changes

Over the performance period of this project, staff changes occurred involving the retirement of one of the planned participants prompting the subsequent inclusion of two other senior staff members. Both staff members were experienced in working with radioactive materials in a fume hood environment while encumbered with personal protective equipment, but they also happened to be inexperienced in XRD specimen preparation methods and more generally in X-ray diffraction methods. With these differences in mind, the associated staff members were incorporated into the study at an earlier point than was originally envisioned. The motivation for this change was twofold. First, it afforded the newly associated staff a highly repetitious opportunity to actively participate in specimen preparation with non-radioactive materials and, secondarily, a similarly repetitious interaction with routine instrument calibration and operation methods. At the same time, the method development study also benefitted from the participation of the two additional staff members as their inclusion allowed reproducibility in the specimen preparation methodology to be evaluated for each participant as well as among participants with different experience levels. Furthermore, with three participants involved, other experimental parameters were also evaluated (e.g., use of a sieve during powder deposition versus not using a sieve) by using the data from one participant as a control and the data from the other two to assess the parameter changed.

# 5. SPECIMEN PREPARATION METHOD DESCRIPTION

## 5.1 Preparation of Saltcake Powder Compounds

As mentioned in section 3, the specimen preparation method selected for development in this study is an established method that is less vulnerable to preferred orientation effects than the packed powder cavity method that it replaced. In general, this method involves applying a thin amorphous layer of adherent to a substrate and subsequently dispersing a finely ground powder form of the analyte material across the adherent layer. The optimization and modification of conventional steps associated with this specimen preparation process were the primary objectives of this project.

While "…grinding sample materials to a 'flour-like' consistency…" is a stipulated step in ATS-LT-507-103, it was decided not to include this step in the method development process. Instead, all of the saltcake simulant materials were initially ground to a fine powder after synthesis was completed in June of 2015 and then reground in January of 2017 after the materials had been transferred to the laboratory where the XRD specimens were to be prepared. In detail, with the exception of natroxalate, by January of 2017 all of the other simulants included significant large agglomerates that had to be re-ground before powder deposition activities could be performed. After the second round of grinding was completed, the finely ground parent saltcake simulant powders were subsampled for single salt specimens and again for each of the binary salt compounds and one last time to prepare the five-salt mixture. For all of the multi-phase mixtures, the parent saltcake phases were combined in a mortar and re-ground a third time - this time to ensure homogeneity of the multi-phase compounds produced. Correspondingly, the powder particle refinement (grind) of each of the deposited powders was held constant among the three preparers. Doing so allowed for other factors (e.g., technique of the individual preparer, deposition method used, material being deposited) to be more clearly evaluated.

## 5.2 Microsieve Design, Construction, and ALARA Features

### 5.2.1 Tool Design

A new tool was designed and constructed specifically for this method development study. One general concern associated with the deposition of tank waste sample powder deposits on an adherent layer relates to controlling the spreadability of the (radioactive) powder during the deposition process. For example, during the powder deposition process the finely ground powder must be transported from the grinding vessel (i.e., mortar) to the substrate for deposition. Given the high airflow in the fume hood environment where the specimens are prepared, airborne spreading of the finely ground powder and subsequent control of the powder during deposition was considered an important method development consideration. In addition, several of the finely ground forms of the saltcake phases under evaluation exhibit a tendency to form loosely bound dry powder agglomerate particles. Some of these agglomerates can be several orders of magnitude larger than the dimensions of the constituent particles. Lastly, in the interest of improved ALARA practice, the tool was intended to be disposable with unused sample material permanently captured inside.

With these considerations in mind, a tool (subsequently referred as a microsieve) was designed and variants were constructed to address the issues raised. Multiple preliminary designs were considered (e.g., a disposable salt shaker prototype) from which the microsieve concept (constructed from two commercially available disposable pipette tips, one being modified with a shaped piece of wire mesh screen permanently adhered in place) was selected for further development.

The initial development work evaluated (1) adhesives used to adhere the wire mesh screen to the modified pipette tip and subsequently (2) the wire mesh sizes to be used during specimen preparation activities. The three adhesives evaluated were (1) Loctite[3] 7649 primer with Loctite 326 adhesive, (2) Permatex[4] 5-minute epoxy (#75157), and (3) J-B Weld[5] plastic bonder.

The Loctite adhesive system failed to cure and therefore failed to adhere the parts. It was interpreted that the adhesive requires all points between the primed surfaces of the parts being joined to come into contact for the adhesive to cure. The open structure of the wire mesh, while thoroughly primed, did not promote curing of the adhesive likely due to the small "primed" contact area of the mesh not catalyzing the adhesive to cure.

The Permatex product information indicates that the epoxy will bond metal and plastic. However, on the reverse side of the packaging, the instructions indicated that the product is not recommended for use on polyethylene or polypropylene. Since this epoxy product did cure but did not bond the wire mesh to the modified pipette tip, it is likely that the pipette is constructed from one of these two common polymers.

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[3] Loctite is a division of Henkel, Düsseldorf, Germany.

[4] Permatex is a division of Illinois Tool Works Inc., Hartford, Connecticut.

[5] J-B Weld, Inc., Sulphur Springs, Texas.

The JB Weld plastic bonder adhesive product was successful in bonding the wire mesh to the pipette tips. Consequently, no further evaluation of other adhesive compounds was undertaken.

As a means of identifying suitable mesh sizes for de-agglomeration of finely ground saltcake powders used in this project, a prototype microsieve stack was constructed using 24-, 30-, 35-, and 40-mesh screens. The range of mesh sizes considered were assessed based upon powder flow through the mesh. The initial evaluation criterion was to identify the finest mesh that passes the majority of the ground powder in a reasonably brief period of time.

Preliminary testing was performed using two tank waste simulants (AN102-2 and AN102 with residual zirconium) due to convenience (i.e., the saltcake simulants were not in the laboratory where the sieves were being constructed). The AN102-2 simulant was ground in a mortar but had a gummy consistency. When placed in the sieve stack, none of the material passed through the 24-mesh screen (see Figure 6). Since this simulant material did not have similar powder flow characteristics to saltcake powders nor was it generally representative of tank waste solids, it was not used in further tests. The AN102 with zirconium-residual simulant was loaded into the sieve stack and processed through as shown in Figure 7. In this case, all of the powder passed through the sequence of wire mesh screens. Subsequently, rather than constructing additional prototype microsieve stacks, 50- and 60-mesh wire cloth was cut into approximately 1-inch square coupons and powder was transferred onto the screen using a microspatula; the screens were then manually oscillated to transmit the powder. Most of the powder passed through both these mesh sizes, although some evidence of size segregation was revealed. As a result of this preliminary testing, two-piece microsieves were constructed using either 40, 50, or 60 mesh wire cloth (aperture dimensions of 420, 297, and 250 microns respectively). The range of mesh sizes selected for microsieve construction considered (1) the simulants tested were not originally part of the test plan, (2) different compounds may have different flow characteristics through the screen meshes, and (3) dozens of specimens were to be prepared, therefore, having tool variants on hand was deemed appropriate.

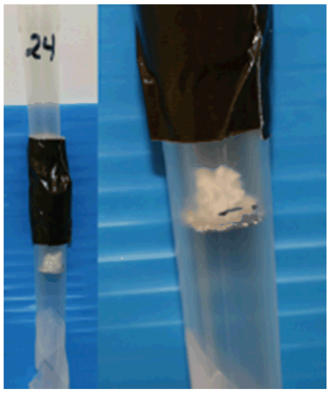

**Figure 6. Prototype sieve stack test failure with a complex tank waste simulant.**

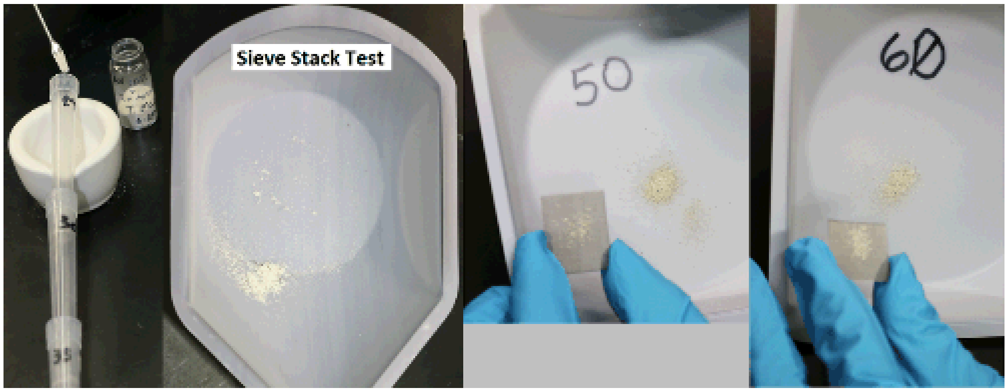

**Figure 7. Wire mesh powder transmission testing is shown in this composite image.**

### 5.2.2 Microsieve Construction

Construction details are shown in Figure 8. Typically, construction involved the following sequence of steps. First, the commercial pipette tips selected (Eppendorf[6] tips, epT.I.P.S, 1-10mL, Cat. No. 022492098) have a line that separates the highly tapered bottom 43mm from the upper section. This cross-section transition was selected as an easily reproducible location for adding the wire mesh. Correspondingly, the lower tip of the pipette was cut off using scissors, then sanded nominally flat and ultrasonically cleaned in a water bath; it was then dried using compressed air.

As mentioned previously, JB Weld plastic bonder was used to adhere the wire mesh to the modified pipette tip, and the components were placed vertically (wire mesh end facing up) in a test tube rack and allowed to dry overnight. Care was taken in the application of the adhesive so as not to block the wire mesh with glue inside the pipette tip. The next day, a handheld power tool (with grinding stone installed), was used to trim the excess wire mesh until the modified pipette fit smoothly and completely into an unmodified pipette (see Figure 8 for both unassembled and assembled versions of the microsieve).

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[6] Eppendorf North America, Hauppauge, New York.

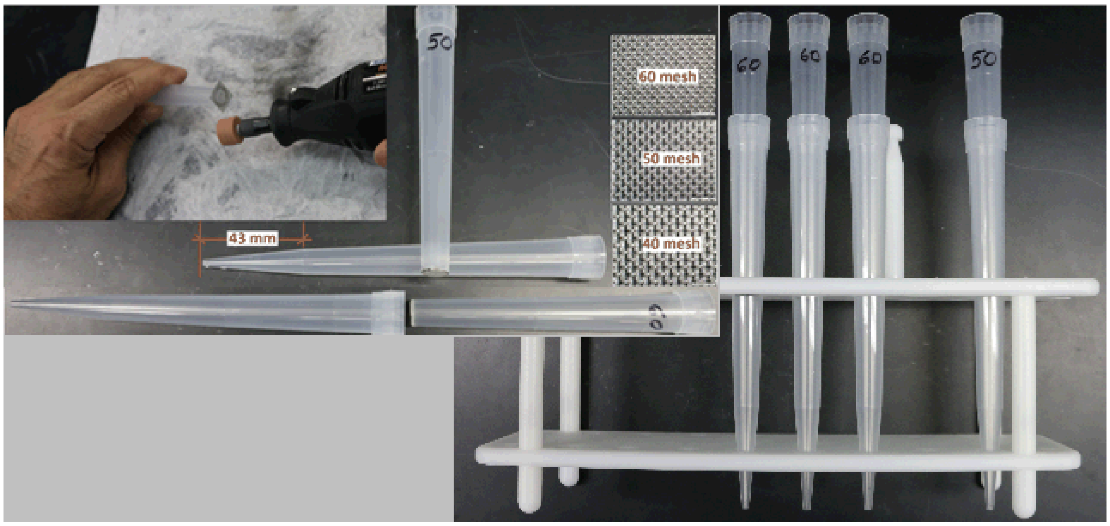


**Figure 8. Construction stages of the microsieves used in this study are shown. (Left Corner Inset) 40/50/60 wire mesh coupon glued to cut end of a disposable pipette prior to trimming. (Main image) Modified pipette sieve trimmed to size ready for insertion into unmodified disposable pipette. (Right, Background Image) Assembled microsieves during construction in a pipette rack which was also used during mesh gluing with glued end facing up.**

### 5.2.3 ALARA Features of the Microsieve Design

The design of the microsieve has several intrinsic features that improve ALARA aspects of specimen preparation. The most obvious improvement is that the powder can be loaded into the tool with it oriented in the horizontal position, thereby capturing all of the powder deep inside the tool. The inlet end of the tool can be plugged with a rubber stopper (i.e., stopper 0, acts both as a powder containment mechanism as well as providing line-of-sight shielding at the inlet) or a piece of tape (powder containment). Next, the mesh screen is located approximately 6.5 cm inside the unmodified pipette tip as shown in Figure 9. The buried depth section means that there are two wall thicknesses of pipette material at and above the cross-section of the tool where the wire mesh screen is located. In the buried region above this, the path length through the tool is further increased geometrically as indicated in Figure 9 and Table 1.

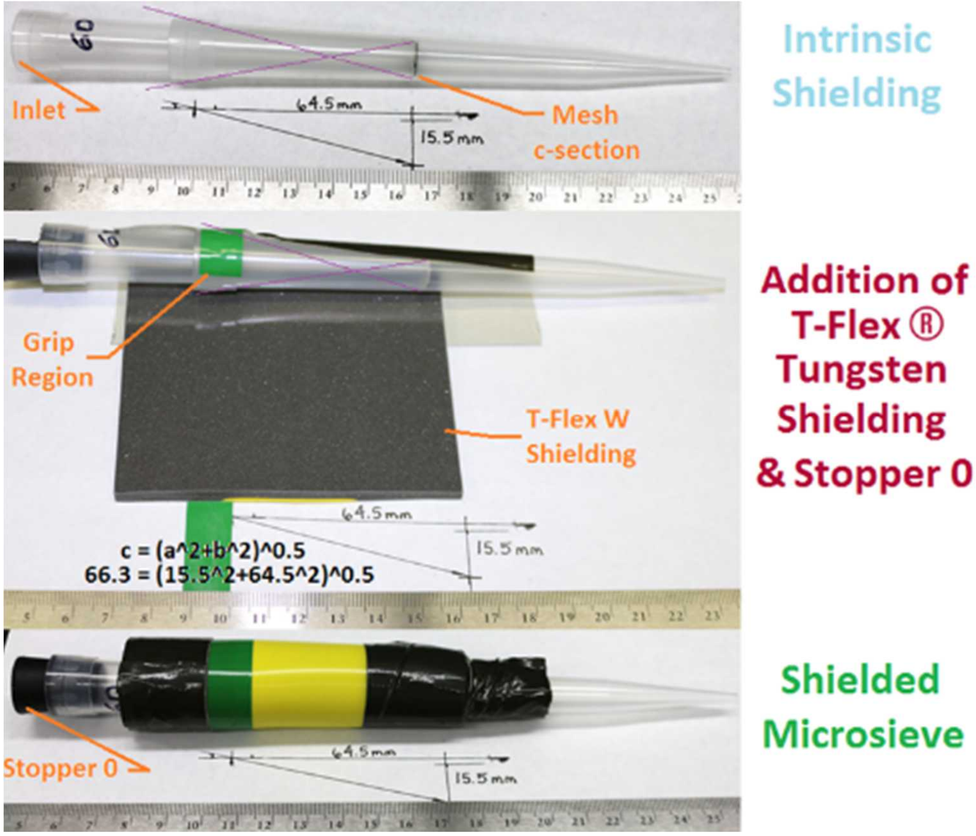
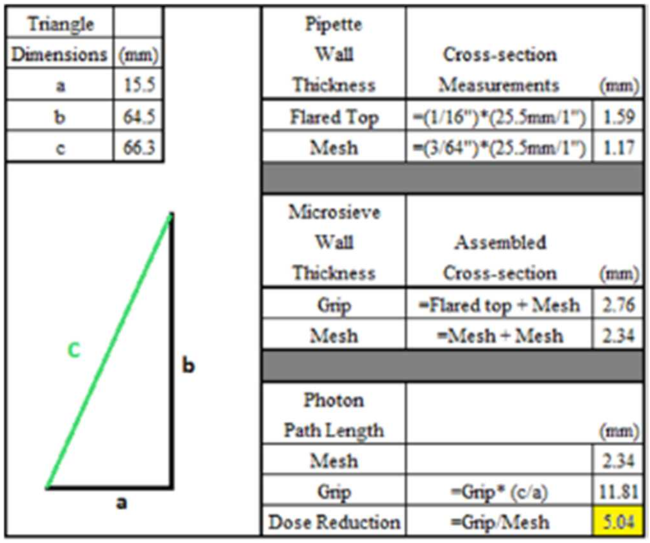


**Figure 9. Several ALARA design features of the microsieve are indicated in this figure. The exit rays highlighted in magenta indicate the minimum degree of inclination (i.e., minimum path length) of exiting radiation through the grip section of the tool.**

**Table 1. Path length calculations of radiation emissions through the wall sections of the microsieve are shown in this table.**



| Triangle Dimensions | (mm) |
|---|---|
| a | 15.5 |
| b | 64.5 |
| c | 66.3 |

| Pipette Wall Thickness | Cross-section Measurements | (mm) |
|---|---|---|
| Flared Top | =(1/16")*(25.5mm/1") | 1.59 |
| Mesh | =(3/64")*(25.5mm/1") | 1.17 |

| Microsieve Wall Thickness | Assembled Cross-section | (mm) |
|---|---|---|
| Grip | =Flared top + Mesh | 2.76 |
| Mesh | =Mesh + Mesh | 2.34 |

| Photon Path Length | | (mm) |
|---|---|---|
| Mesh | | 2.34 |
| Grip | =Grip* (c/a) | 11.81 |
| Dose Reduction | =Grip/Mesh | 5.04 |

Here for sake of mathematical simplicity, a right triangle whose elevation (i.e. "b" dimension in Table 1) was determined by the mesh location and the center of the "grip region," and the base of the triangle was defined by the mesh cross-section (i.e., "a" dimension in Table 1). The inclination angle depicted connects the grip region with the far side of the mesh yielding the shortest path length to the user's fingers. This is intended to represent the worst case (i.e., highest received dose) scenario. It is acknowledged that the fingers holding the tool occupy a vertical portion of the device, and therefore the calculated results are to be considered indicative rather than precise.

The grip region of the tool was selected/specified based upon (1) leverage/tool control, (2) distance – between the user's fingers and the sample materials (particularly beneficial for radioactive materials), and (3) shielding – this is the thickest cross-section of the microsieve. Points 2 and 3 are key elements in addressing ALARA practice. In particular, the grip region of the lower pipette is 4/3 the thickness of the lower pipette wall cross-section. The geometry illustrated in Table 1 compares the relative shielding at the mesh cross-section with the corresponding shielding in the grip section. By geometry, the inclination angle between the sample material and the grip section further elongates the photonic path length radiation would have to traverse to reach the user's fingers. Thus, it was calculated that a relative dose reduction of approximately 5 times would be achieved by holding the tool in the grip region rather than at the mesh cross-section and this was without considering geometric spreading of the emitted radiation.

In some future instances it is foreseeable that the intrinsic shielding properties of this tool may not be sufficient. However, the design of the tool easily allows for the addition of formal shielding materials as shown in the lower portion of Figure 9. Shielding benefit calculations similar to the ones presented in Table 1 can be performed using the T-Flex[7] gamma-ray shielding product.

After deposition activities have been completed, the microsieves are intended to be disposed of with contents retained. This addresses the "time" aspect of good ALARA practice (i.e., time, distance, and shielding). Here the concept is that the tool does not need to be cleaned or emptied of contents. This disposability aspect of the design helps minimize the contact time with the device user.

Final disposition involves applying tape or Parafilm M[8] (at minimum) to both ends of the microsieve. At the deposition end, the tip end can be placed directly on a prepositioned piece of tape. Next a pair of tweezers (typically 6 inches or longer) can then be used to grab the free end of the tape and fold it over the top surface of the end of the microsieve being sealed. The inlet end of the tool can be sealed using similar methods (i.e., if a rubber stopper was not used). The sealed tool and contents then can be transferred from the work space to the microsieve waste container (see Appendix I, Figure AI-1).

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[7] T-Flex is a trademark of Eichrom Technologies LLC, Lisle, Illinois.

[8] Parafilm M is a trademark of Bemis Company, Incorporated, Oshkosh, Wisconsink

### 5.3 Specimen Preparation Tools

The collection of tools used in this project for XRD specimen preparation method development are shown in Figure 10. A ½-inch hole punch was also used but is not shown in Figure 10.

**Figure 10. All of the tools, specimen supports, and associated ancillary components used to prepare XRD specimens are shown in this figure.**

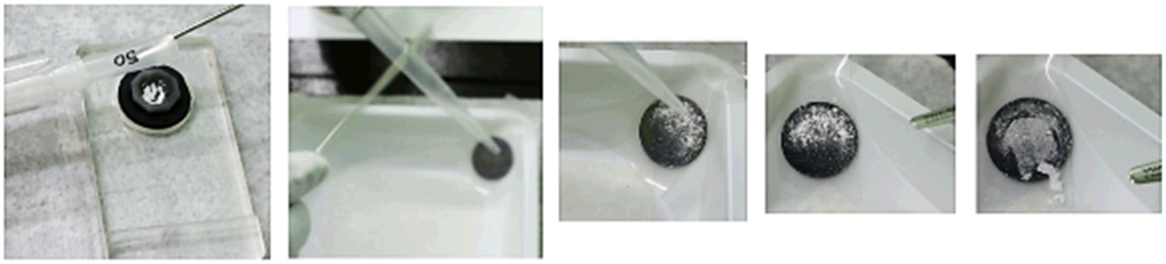
### 5.4 XRD Specimen Preparation Sequence

The step-by-step sequence used to prepare XRD specimens is documented in Figures 11-13.

**Figure 11. The specimen preparation sequence is illustrated starting with the top left proceeding to the right, then continuing on the bottom row, left to right.**

Apply Powder Tap Weigh Boat Uniform Coating

**Figure 12. Method I - Powder deposition using a microspatula is illustrated in this composite figure advancing from left to right.**



Load Sieve Tap Sieve Coat Top Tap Weigh Boat Inspect Coating

**Figure 13. Method II - Powder deposition using a microsieve is illustrated in this composite figure advancing from left to right.**

The first action required involves preparing a mask for the single crystal substrate. The mask is made of a piece of low adhesive tape (PATCO 5560[9], Removable Protective Film Tape) with a centered hole punched in it. A length of approximately 1.5 to 2.0 inches is cut and applied to a piece of release paper. The excess length beyond the 1-inch substrate dimension is folded back on itself to make a tab that will be used to assist in removal of the mask after powder deposition activities are complete. Then the ½-inch hole punch is used to generate a centered hole in the mask.

The descriptions that follow are matched to the image sequences which generally proceed from the top left to bottom right (see Figure 11). The release paper is removed and the mask is applied to the surface of the single crystal silicon substrate. A cotton swab then is used to fully adhere the mask to the surface, paying particular interest to the inner edge of the mask to ensure that none of the adherent bleeds under the mask during subsequent adherent melting. Continuing, petroleum jelly is applied to the unmasked portion of the silicon substrate, then a glass slide is used to screed off the excess leaving a uniform, thin layer of petroleum jelly on the unmasked portion of the substrate. In the interest of generating a reproducibly flat surface for powder deposition and also to promote the adhesive and compliance properties of the petroleum jelly, the substrate typically is heated to approximately 90 ºF to liquefy the jelly. Once completely melted, the substrate is carefully removed from the heat source and allowed to re-solidify. The still warm adherent layer is more compliant than cold petroleum jelly, which allows high aspect ratio particles to more successfully implant thereby enhancing the randomness of the particle orientations in the deposited powder. The substrate is then placed in the corner of a weigh boat ensuring that the deposition surface of the substrate is inclined. Powder deposition then proceeds using one of two developed methods.

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[9] PATCO 5560 is manufactured by Berry Plastics Corporation, Franklin, Massachusetts.

In method I (see Figure 12), finely ground sample material in transferred to the substrate just above the unmasked region using a microspatula or other equivalent tool (see Figure 12). The powder is dispersed across the adherent layer by tapping on the edge of the weigh boat or the substrate itself. This action causes the powder to tumble across the adherent layer as a means of randomizing the orientation of the powder particles.

Alternately in method II (see Figure 13), a microsieve is loaded with a small volume of powder, then the tip of the microsieve is placed above the unmasked portion of the substrate (see Figure 13). The microsieve is then tapped on using another tool (e.g., clean microspatula). This tapping action causes powder to pass through the wire mesh screen and exit the microsieve. Once a reasonable amount has been deposited near the top of the substrate, the substrate and/or weigh boat is tapped to distribute the powder across the surface of the substrate.

Common to both powder deposition methods is the process of removing excess powder not adhered directly to the petroleum jelly (see bottom row of Figure 11). The excess is tapped off of the substrate. Typically, the deposit is then inspected, and more powder is distributed as previously described on an “as-needed” basis until a uniform powder deposit has been produced.

Next, the mask is carefully removed using tweezers to pull the mask while the wood stick end of a cotton swab is used to hold the substrate down while removing the mask. Normal incidence is recommended when using the wood end of the swab to hold the substrate while removing the mask. The surface of the mask can be slippery due to exposure to the petroleum jelly. Normal incidence maximizes the contact area and minimizes the opportunity for the wood tool to slip and mar the powder deposit.

Subsequently, plastic tweezers are used to seat the substrate (deposit facing upward) in the small quantity of petroleum jelly adherent. The purpose of the small amount of petroleum jelly in the cup is twofold. First, the petroleum jelly minimizes the opportunity for the substrate to accidentally come out of the cup before measurements are complete. Second, the adherent also minimizes damage to the substrate during measurements performed while the specimen is spinning (spinning specimens during measurements helps to improve the randomness of particle orientations during diffraction measurements but also can cause the silicon substrate to rattle around in the aluminum sample cup). However, applying too much petroleum jelly in the cup can displace the substrate resulting in specimen surface displacement errors, and the excess adherent also makes it significantly more difficult to remove the substrate for cleaning after the measurements are complete. At this point, the specimen preparation process is complete and the specimen is available for transport to instruments (optical microscopy and XRD).

## 5.5 Discussion of Saltcake Specimen Preparation Tools and Techniques Evaluated

### 5.5.1 Mask Material Evaluation

Scotch brand 1-inch wide, clear packing tape; and Patco 5560 removable protective film tape were preliminarily evaluated in this study as candidate mask materials. The Scotch tape resulted in the cleanest edge cuts but proved to have too strong an adhesive, making it difficult to remove the mask after powder deposition and frequently resulting in tool marks (i.e., evidence of tool slippage) being left in the deposit.

Packing tape was also found to be unsuitable as a mask material for two reasons. First, the thickness of this type of tape resulted in poor quality cut edges on the perimeter of the unmasked region. These edge irregularities frequently resulted in petroleum jelly leaking under the mask during the melting (i.e., deposit self-leveling) step. Second, this tape's adhesive is very strong making it very difficult to remove the mask after the sample material has been deposited, resulting in similar and frequent deposit artifacts.

The Patco 5560 tape was the best mask material identified in this study. This mask material is thicker than the Scotch tape and does occasionally result in adhesive artifacts along the cut perimeter of the mask. For clarity, the Patco 5560 adhesive artifacts observed to date typically occur on the exterior edges of the petroleum jelly deposits and include small irregular absences (typically less than 0.5 mm) of petroleum jelly adherent. When the mask was removed (i.e., after powder deposition), the edge irregularities were evident as the adhesive residue always departs with the removal of the mask. These edge effects did not have a significant impact on the overall quality of the diffraction as the affected fraction of the deposit typically represented less than 1% of the entire deposit area.

The relative ease in cutting the hole in this tape, the ease of sealing it to the substrate, the chemical compatibility with the petroleum jelly, and the ease of removal after the deposit has been formed resulted in this product being selected as the only mask material used for all of the specimen preparations discussed in subsequent sections of this report.

### 5.5.2 Petroleum Jelly Adherent Evaluation

Two petroleum jelly brands (Vaseline™[10] and Equate™[11]) were initially tested. No observable differences were found in the diffraction data from either brand. However, the older jar (Vaseline) was discarded early in the test plan as a significant amount of visible debris was present in the source container. Thus, in the interest of not introducing foreign debris effects into the specimen preparation test results, all of the work reported was performed using the new jar of Equate brand petroleum jelly.

### 5.5.3 Contamination Control Mechanisms

A large format (14cm x 14cm) plastic weigh boat was placed on a (32cm x 32cm) moistened paper towel thereby defining the work space on the floor of the hood with the exact positioning as determined by the individual preparing the specimen (see Appendix I, Figure AI-1). By procedural design (see section 5.4, Figure 11), the weigh boat was intended to be the primary vessel used to control the spread of powdered sample material within the fume hood environment. The paper towel on the floor of the hood was a secondary control mechanism. During the powder deposition process, as visible powder accumulated in the bottom of the weigh boat, small coupons (roughly 8cm x 8cm) of damp paper towels were intermittently added to the

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[10] Vaseline is a trademark of Chesebrough Manufacturing Company, New York, New York.

[11] Equate is a trademark of Wal-Mart Stores, Inc., Bentonville, Arkansas.

bottom of the weigh boat using tweezers. These damp paper coupons trapped loose powder under them while also providing a clean/new work surface for actions (e.g., removal of excess powder on the deposit, inspection of the deposit, mask removal, wiping the bottom of the substrate before transferring the substrate into the sample cup in case the bottom was contaminated during powder deposition). Since the specimens evaluated within the context of this study were all non-radioactive, the success of these controls was not proven. However, other studies involving radioactive materials carried out over the same time interval used the same contamination controls and spreadable contamination control was successfully demonstrated.

### 5.5.4 Microspatula Evaluations

Various microspatulas were used and evaluated in the course of this study. Personal preference varied somewhat from individual to individual with long (VWR[12] #82027-526, 16.5 cm long) and short (Fisher Scientific[13] #2140115, 14 cm long) handle microspatulas being used interchangeably by the participants of this study. However, best (i.e., ALARA) practice suggests that the long handle versions of the tool are preferred for direct interaction with radioactive materials during the powder deposition process due to increased working distance between the sample material and the preparer. At the same time, the shorter version of the tool was found to be clearly distinguishable from the long version making it easy to maintain the short version as “clean” for tapping processes (non-contact uses). Since microspatulas were used for multiple tasks, the working practice of segregating “in-use” tools, where preparers distinguished between sample-contacted versus non-contacted tools, was used throughout this project. Individual tool selection and use practice was left to the preferences of the individual participants throughout this study.

### 5.5.5 Tweezers Evaluations

Generally speaking, short (i.e., 10 - 13 cm length) plastic tweezers were preferred for handling the single crystal silicon substrates directly rather than their metal counterparts because the plastic versions did not easily scratch or otherwise damage the silicon. At the same time, similarly sized metal tweezers were much more successful in the mask removal step due to the metal tweezers being much more flexurally stiff. Metal tweezers with polymer tubing over the working ends proved to be a viable compromise version of the tool useful for all aspects of specimen preparation. This modification was both stiff enough to peel the mask off after the deposit was formed but also soft enough at the contact points when handling the single crystal silicon substrate that no damage occurred. In spite of this general utility, “clean” and “sample contacted” tweezers were used during the various specimen preparations steps. It was possible to clean one pair of tweezers in-process, however, this practice was not considered to be best ALARA practice for working with radioactive sample materials as the extra time involved with repeated cleaning operations increases “exposure” time in a relatively small space (i.e., fume hood) when radioactive materials are present.

For cleaning applications (excluding interactions with the silicon substrate), longer format (i.e., 20 – 25 cm in length) metal tweezers were exclusively used. These were wrapped with damp

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[12] VWR International, Radnor, Pennsylvania.

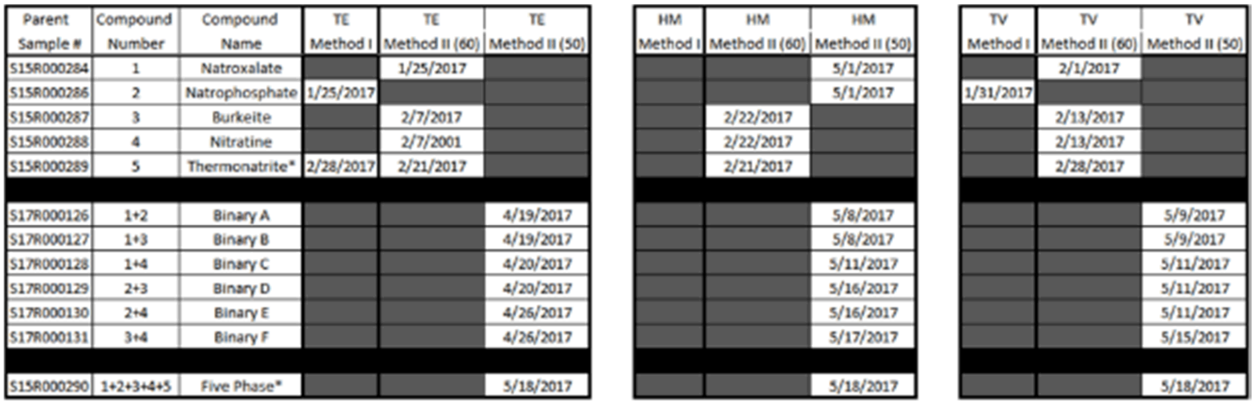
[13] Fisher Scientific is a brand of Thermo Fisher Scientific Inc., Waltham, Massachusetts.

paper towel coupons for the purpose of wiping off potentially contaminated parts while simultaneously providing greater distance between the hands of the individual performing cleaning operations and the sample contacted parts being cleaned. Similar dimension plastic tweezers were not tested as there was not an obvious "cleaning activity" advantage associated with them. Ultimately, the decision of which hand tools were used was left to the judgment of the preparer of each specimen during this project.

### 5.5.6 Deposition Method Evaluation

Sample powder deposition via Method I & Method II as described in section 5.4 above were used to prepare specimens as indicated in Table 2 below.

**Table 2. Summary of the powder deposition methods used and corresponding data acquisition dates. Each cell represents (3) specimen preparations and XRD data sets.**



| Parent Sample # | Compound Number | Compound Name | TE Method I | TE Method II (60) | TE Method II (50) | HM Method I | HM Method II (60) | HM Method II (50) | TV Method I | TV Method II (60) | TV Method II (50) |
|---|---|---|---|---|---|---|---|---|---|---|---|
| S15R000284 | 1 | Natroxalate | | 1/25/2017 | | | | 5/1/2017 | | 2/1/2017 | |
| S15R000286 | 2 | Natrophosphate | 1/25/2017 | | | | | 5/1/2017 | 1/31/2017 | | |
| S15R000287 | 3 | Burkeite | | 2/7/2017 | | | 2/22/2017 | | | 2/13/2017 | |
| S15R000288 | 4 | Nitratine | | 2/7/2001 | | | 2/22/2017 | | | 2/13/2017 | |
| S15R000289 | 5 | Thermonatrite* | 2/28/2017 | 2/21/2017 | | | 2/21/2017 | | | 2/28/2017 | |
| | | | | | | | | | | | |
| S17R000126 | 1+2 | Binary A | | | 4/19/2017 | | | 5/8/2017 | | | 5/9/2017 |
| S17R000127 | 1+3 | Binary B | | | 4/19/2017 | | | 5/8/2017 | | | 5/9/2017 |
| S17R000128 | 1+4 | Binary C | | | 4/20/2017 | | | 5/11/2017 | | | 5/11/2017 |
| S17R000129 | 2+3 | Binary D | | | 4/20/2017 | | | 5/16/2017 | | | 5/11/2017 |
| S17R000130 | 2+4 | Binary E | | | 4/26/2017 | | | 5/16/2017 | | | 5/11/2017 |
| S17R000131 | 3+4 | Binary F | | | 4/26/2017 | | | 5/17/2017 | | | 5/15/2017 |
| | | | | | | | | | | | |
| S15R000290 | 1+2+3+4+5 | Five Phase* | | | 5/18/2017 | | | 5/18/2017 | | | 5/18/2017 |

* not phase pure

Method I was found to be relatively easy to perform; however, this technique does have several drawbacks. First, due to the airflow in the fume hood environment where the specimens were prepared, transferring powder from the source vessel (sample container in this study but more generally the point of origin would be a mortar) to the masked substrate can result in powder being spread between the two end points. Second, finely ground saltcake powder tends not to depart the microspatula as individual particles but often as large loosely bound agglomerates. Third, since the amount transferred to the deposition surface is difficult to control (due to formation of loosely bound dry agglomerates), a larger volume of sample material tends to be used per specimen with this powder deposition method.

Method II clearly affords the preparer more control over spreadability of the powder. Beyond the limited domain of this study in cases where the hazard level is elevated, the inlet end of the microsieve could be sealed (e.g., tape, or rubber plug if more shielding is needed). However, this was infrequently performed in the course of this project. In general, the microsieve allowed for very reproducible specimens. However, the 50-mesh version was clearly preferred due to better powder transmission characteristics. To be clear, the 50-mesh microsieves passed the powder material quickly through the screen resulting in powder deposits that could be completed in approximately 30 seconds. Thus, from an ALARA perspective, 50 mesh was clearly preferred. From a data quality perspective, the 50-mesh microsieves also yielded a denser population of powder particles on the adherent layer.

On review of the data, the 60-mesh version of the microsieve was found to be too restrictive with substantial differences from one saltcake compound to the next, resulting in very thin sparse powder deposits in the worst cases that took an unacceptably long time to deposit (beyond ten minutes).

Both Method I and Method II will continue to be used as the details of individual specimen preparation (e.g., powder properties, powder volume) in conjunction with the experience of the preparer dictate. In analyses performed outside of this project, the microsieves used in Method II were found to be problematic in the processing of magnetized materials due to attraction to the wire mesh cloth used internally (RPP-RPT-60141, *Report on the Physical and Chemical Analysis of Solids Recovered from the Robotic Crawler Deployed to the Annulus of Tank 241-AZ-101*). In future cases involving similar sample materials, Method I deposition is recommended.

Static charging of the microsieves was also found to be a problem, making it difficult to load powder into the microsieve. This issue was successfully mitigated, however, by coating the inside and outside of the tool with an anti-static solution and allowing the microsieve to dry overnight.

### 5.5.7 Deposition Technique Evaluation Using the Microsieve

#### 5.5.7.1 Microsieve Orientation

The horizontal to vertical inclination of the microsieve did impact the volume of powder emanating from the deposition end. This could be readily adjusted by the user depending upon how the powder material interacted with the microsieve. Among the saltcake simulants tested, some compounds flowed more freely through the mesh while others required significantly more tapping effort and more vertical orientation to pass enough material to generate a uniform deposit layer.

#### 5.5.7.2 Powder Positioning During Deposition

Deposition at the top of the substrate was described in section 4.4 with subsequent powder distribution being achieved via tapping on the weigh boat and/or substrate. However, rastering the tip of the microsieve to control exactly where powder was deposited on the petroleum jelly adherent layer was also performed many times during this method development study. While both yielded uniform powder deposits, the rastering method definitely took longer with no obvious difference in the data quality. Here, competing best ALARA advantages were considered. The rastering method definitely minimizes the amount of material delivered to the specimen resulting in optimum ALARA practice with regard to the issue of spreadability of radioactive contamination. The ALARA counterpoint considered and also deemed most significant, was the “top of substrate tap to distribute” approach. This approach generally involved more powder being delivered to the substrate. However, the resulting deposit was formed in seconds (with practice) compared to minutes with the rastering method. Minimizing exposure time and the associated dose received by the preparer, especially when working with radioactive materials, was considered to be the more important benefit. To make the point more clearly, other supplemental "spreadability limiting control mechanisms" (e.g., preparing specimens inside a bag within the hood environment) were, and continue to be, available on an as-needed basis. At the same time, alternate "time reduction" techniques were not obvious.

### 5.5.8 Use of Plastic Components in Specimen Preparation

Finely ground saltcake compounds inadvertently dispersed on occasion due to the presence of static charge on the surface of plastic parts associated with specimen preparation activities. Weigh boats, microsieves, tweezers, mortar supports, and petri dishes (used for specimen transport from the fume hood environment to instruments for measurements) were all components that were observed to scatter ground powder materials both within and beyond the domain of this study. Most of these parts can be wiped with a commercially produced anti-static product (e.g., ALC Staticide[14], #2010) to alleviate this problem.

## 5.6 Advantages and Limitations of the Thin Powder Deposition Specimen Preparation Method

### 5.6.1 Advantages

Several clear advantages of the thin powder method relative to the packed-powder cavity method were identified. First and most significantly, the primary objective of the test plan (i.e., substantially minimizing if not entirely eliminating deleterious preferred orientation effects in the diffraction data from saltcake forms of tank waste materials evaluated for solid phase content) was addressed by selecting the thin powder specimen preparation method for further development. The successfulness of this method will be discussed in section 7 and demonstrated in the data included in Appendices I-III.

Second, the volume of material used to generate the mount was vastly reduced (approaching two orders of magnitude of volume reduction). This volume reduction has an associated radiation exposure reduction at every step in the specimen preparation process. Correspondingly, while both preparation methods bind the powder in some form (petroleum jelly adherent for thin powder mounts versus collodion binder in packed powder mounts), the potential for accidental specimen drops inside or outside the fume hood always exists. Given the smaller total volume of material involved, the risk of accidental spreading of sample material is significantly reduced (i.e., the thin powder deposit specimen preparation method is the better ALARA practice).

Third, for an experienced preparer, the sample preparation times were comparable between the two methods (using past experience preparing cavity mounts with the thin powder mounts evaluated in this study). However, the post-measurement cleanup time and associated exposure to radiation are not comparable for the two specimen mounts. In the substrate clean-up processes (where the differences were most pronounced), thin powder mounts were inverted and placed on a damp paper towel and manipulated with long tweezers to remove all visible sample material

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[14] ACL Staticide is a product of ACL, Inc., Chicago, Illinois.

from the deposit surface. Using this deposit clean-up method, the substrate itself acted to shield the individual performing clean-up operations. Subsequently, the substrate was ultrasonically cleaned to remove microscopic sample material residuals. The total exposure time for the person performing clean-up activities was typically less than one minute.

On the other hand, the process to remove mounted, packed powder material that was routinely bound with collodion has typically required dissolution (e.g., using ethanol) prior to removal from the cavity. One of the more time consuming aspects of this process has involved extracting bound material from the 90 degree corners at the base of the cavity. Typically, multiple wet wiping operations (ethanol and/or water) have been required to achieve an equivalent condition of "no visible" sample material being present prior to ultrasonic cleaning. This clean-up process can take several minutes and sometimes is less than 100% effective in removing all of the material bound in the corners.

### 5.6.2 Limitations

One significant limitation of the thin powder mount method relates directly to the use of the petroleum jelly adherent. As indicated in Figure 3, the adherent does have a significant scattering signature, and the presence of two crystalline peaks can overlap with the diffraction response of some of the constituent analyte phases present in the data as well. For instruments with grazing incidence measurement capability (the Rigaku MiniFlex II used in this method development study does not have this capability), the significance to the adherent layer can be successfully diminished by selection of a shallow incidence angle. For the work covered in this report and continuing XRD-based evaluations of solid phase content using the MiniFlex II, the petroleum jelly adherent did and will continue to obscure the presence of amorphous content in the sample powder deposited on the petroleum jelly adherent.

### 5.7 Additional Specimen Preparation Tools and Associated Topics

Additional specimen preparation tools and ALARA improvement techniques associated with the generation of XRD specimen mounts and their post-analysis disposal have been developed but not as a direct part of this method development study. The details associated with the development of contamination area fume hood XRD specimen preparation tools are described in Appendix IV.

## 6. DATA PROCESSING

Typically diffraction data were processed to evaluate the crystalline component(s) of powder form analyte materials in this project and, more generally, for tank waste materials using a standard sequence of data processing steps. First, the data file was opened in MDI Jade 9.7. Next, a background shape model was specified by the analyst using a spline curve. Subsequently, the search/match algorithms in Jade were invoked, and the International Centre for Diffraction Data Powder Diffraction File (PDF 4+) was accessed to compare the raw data with background model to reference information stored in the PDF 4+ database. Typically, the entire database was searched first with subsequent searches performed as needed. Secondary searches targeted smaller subfiles (e.g., Inorganic subfile, Minerals subfile) in the database and/or added search filters (e.g., adding elemental chemistry defined by energy dispersive spectrometry) until most if not all of the diffraction peaks were associated with a known phase. For the 108 data files acquired in this study, unidentified peaks were rare occurrences prior to evaluating the diffraction data from the five-phase mixture specimens. For this powdered compound, it was deemed likely that peak interferences involving the two unintended phases (i.e., trace levels of trona and nitratine were present in the mixture) and the five primary phases of interest resulted in incomplete numerical deconvolution during the database search process, resulting in a few unidentified peaks.

# 7. RESULTS

## 7.1 Single Saltcake Phase Results

### 7.1.1 Diffraction Pattern Repeatability

On review of the diffraction patterns in Appendix I (see Figures AI-1, AI-3, AI-5, AI-7, and AI-9), the patterns are exceptionally similar. It should be mentioned that the 9 individual diffraction traces in each plot were offset in equal intensity increments to make pattern comparisons easier. No systematic differences were observed in the data based upon who prepared each specimen or which powder deposition method was used.

### 7.1.2 Phase Identification Results

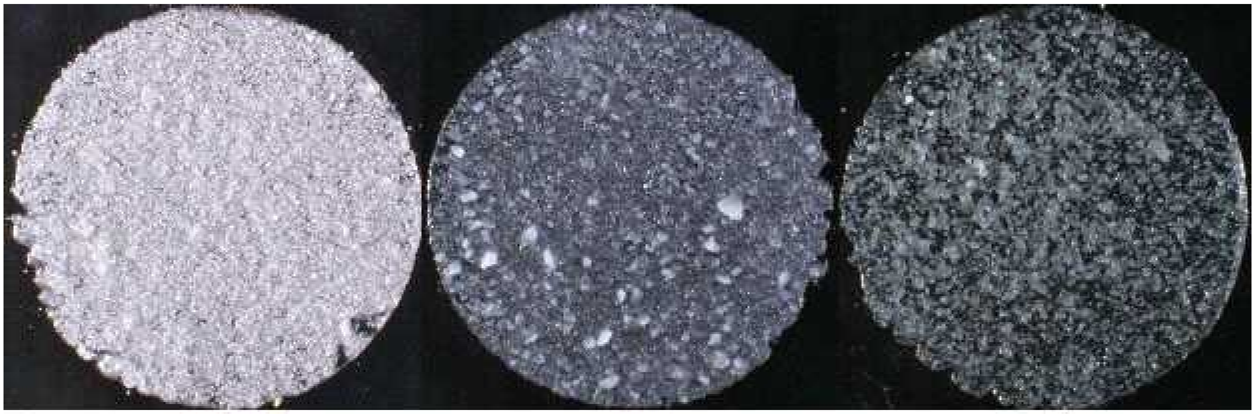
For all of the specimens prepared (3 individuals x 3 repeat preparations) using the first four saltcake simulant materials (i.e., natroxalate, natrophosphate, burkeite, and nitratine), the analyte saltcake phase was successfully identified. In the case of the fifth saltcake simulant, the intended simulant phase (i.e., thermonatrite) was correctly identified in addition to two other unintended phases (trona and nitratine). The presence of significant quantities of trona and nitratine suggest the amount of sodium hydroxide added to the stock solution portion of the simulant recipe during synthesis was not correct.

### 7.1.3 Specimen Micrographs

On review of the specimen images in Appendix I (see Figures AI-2, AI-4, AI-6, AI-8, and AI-10), seemingly significant differences do appear. Given the diffraction-based results reported in the previous two subsections (7.1.1 and 7.1.2), the apparent differences in specimen deposit appearance did not have an observable impact on the phase content analysis. That being said, some of the differences are traceable to when the images were taken (i.e., before the diffraction measurements or after). Some of the differences are traceable to the powder deposition method used in conjunction with the material being deposited. For example, all nine natroxalate deposits tended to be dense while all nine thermonatrite deposits tended to be more sparsely populated powder distributions. This behavior is indicative of the agglomeration properties of each of the powders and how those properties interacted with the 60 mesh-screens used in the microsieves involved with each of these specimens. In this stage of the project, all of the participants found the 60-mesh version of the microsieve difficult to use when preparing the burkeite, nitratine, and thermonatrite specimens. These difficulties strongly influenced the decision to switch to the 50-mesh version of the microsieve for all of the saltcake mixture specimen preparations. Finally, material-based agglomeration differences were also influential in the appearance of the specimen deposits. Burkeite and nitratine both proved capable of forming an agglomerate after passing through the mesh and exiting the lower portion of the microsieve in loosely bound agglomerate form. This tendency seem to be enhanced when the microsieve was oriented closer to a horizontal orientation. It is recommended practice to orient the microsieves vertically, only tilting the microsieve to control the powder flow rate out of the tool.

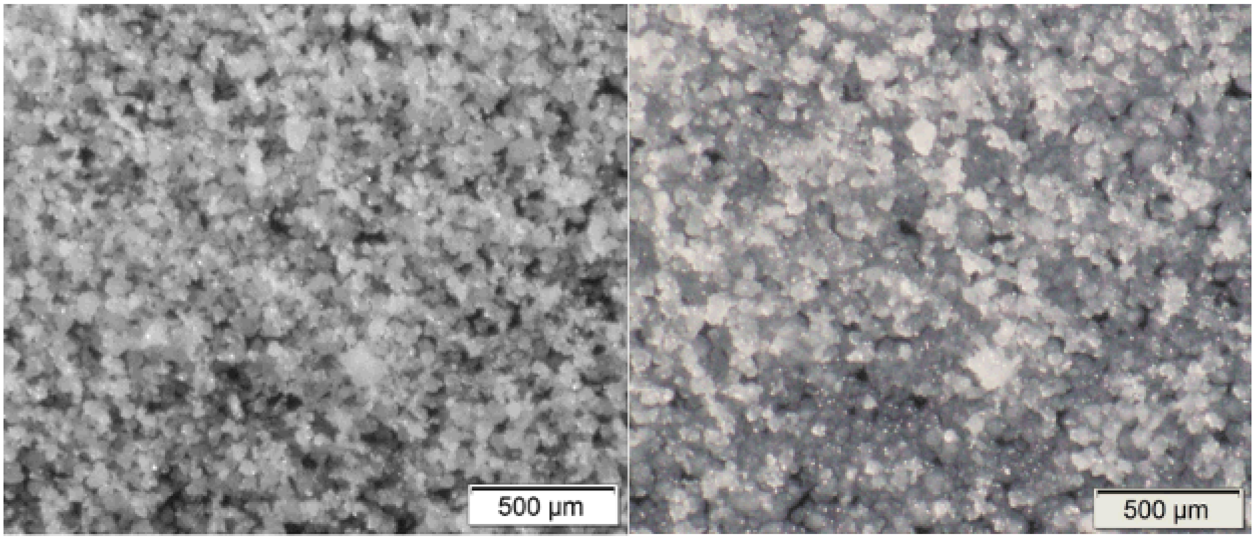
### 7.1.4 Adhered Powder Deposit Transformation into Contiguous Crust

Early in the project the specimen micrographs were taken after the diffraction measurements were completed. While the micrographs were being acquired, it was noticed that the powder deposits had changed in appearance. The deposited materials had an icy translucent appearance relative to the original snow-like white powder deposit (see Figure 14). As indicated in the figure, this clearly was not a material specific process. This transformation of powder into contiguous crust happened to every specimen produced regardless of who prepared the specimen, what saltcake simulant was deposited, or which method was used to deposit the powder. To further investigate this phenomenon, a new natroxalate specimen was prepared and time lapse images were acquired (see Figure 15). It should be mentioned that the powder-to-crust transformation process was observed happening very much more rapidly in the warmer months of this project (see Figure 16). Although not incorporated in this report, microscope-based movies of the transformation process were acquired later in the project (i.e., when ambient room temperatures were higher in the laboratory). In these movies, the powder to crust transformation process was completed in as few as 5 minutes following deposit of the powder. This transformation process was not specific to saltcake materials. Beyond the scope of this project, a similar transformation process has been informally documented for tank waste materials as well as for various forms of cancrinite.



Natroxalate Burkeite Thermonatrite

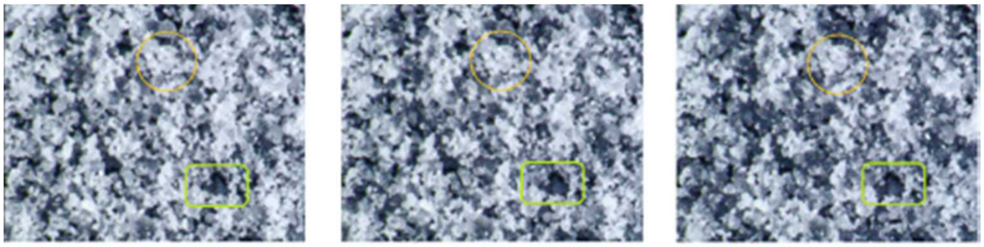
**Figure 14. Transformed specimen deposits for three saltcake simulants prepared by the same individual using the same powder deposition method (Method II 60 mesh) are shown in this composite image.**



Time = 0 minutes, Natroxalate

Time = 180 minutes, Natroxalate

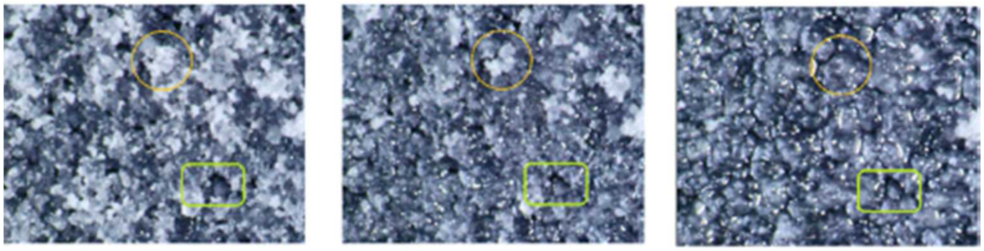
**Figure 15. The left image of this figure captured the powder deposit morphology within a few minutes of when the powder was deposited on the adherent. The right image shows the same deposit 3 hours later, after the powder partially transformed into a contiguous crust.**



As Deposited

Time = 30 seconds

Time = 60 seconds



Time = 90 seconds

Time = 120 seconds

Time = 150 seconds

**Figure 16. Time lapse images were captured from a movie of binary compound A (50:50 mixture of natroxalate & natrophosphate). The yellow circle and green rectangle highlight features that change during the transformation process.**

One unintended benefit of this powder deposit transformation is that the fused contiguous crust form of the deposit substantially diminishes the risk of inadvertently spreading contamination relative to the powdered form.

For sake of clarity it should also be noted that the diffraction data were reviewed for evidence of preferred orientation effects resulting from the transformation process. None could be identified.

### 7.1.5 Mass Loss Associated with the Powder Transformation Process

A limited effort to further understand the powder transformation process was undertaken. A deposit of petroleum jelly and natroxalate was placed in the bottom of a weigh boat. The weigh boat with deposit was placed on a shielded balance with the doors closed, and the weight was monitored as a function of time. Similar measurements were made with just petroleum jelly in an identical weigh boat and just natroxalate powder in yet another identical weigh boat. Only the powder deposit weigh boat yielded systematic evidence of a reaction involving weight loss. For the sake of completeness, the aluminum sample cup and single crystal silicon substrate typically involved in XRD specimen preparation were not used in this experiment in the interest of maximizing the weight sensitivity of the reported weight measurements. The balance sensitivity was verified prior to the onset of this experiment in accord with ATS-LO-140-008, "Routine Use and Quality Assurance for Analytical Balances at 222-S Laboratory Complex." The corresponding weight loss data are presented in Figure 17. An unambiguous declining trend is evident. It is not clear whether the apparent out gassing is associated with decomposition of the petroleum jelly or the deposited powder. Although this was an interesting result, no further effort was undertaken during the course of this project as this transformation process does not appear to have a negative impact on the quality of diffraction data generated.

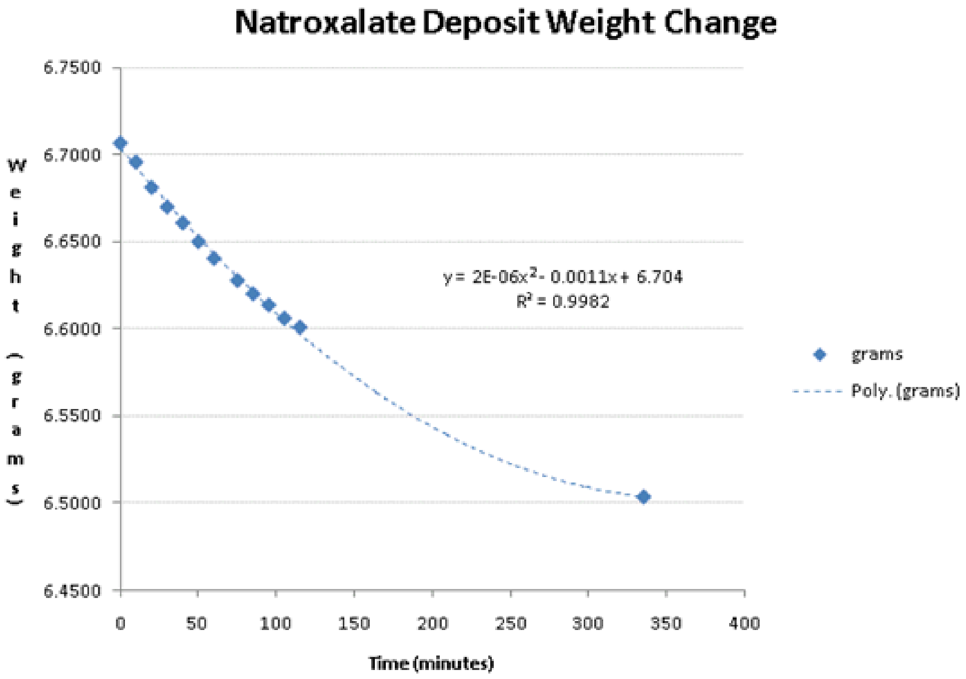


**Figure 17. Weight loss data as a function of time for a natroxalate powder deposit on petroleum jelly applied to the surface of a weight boat similar to the methodology described in section 5.4 is shown in this plot.**

## 7.2 Binary Saltcake Compound Results

### 7.2.1 Diffraction Pattern Repeatability

On review of the diffraction patterns in Appendix II (see Figures AII-1, AII-3, AII-5, AII-7, and AII-9), the patterns are exceptionally similar. It should be mentioned that the nine individual diffraction traces in each plot were offset in equal intensity increments to make pattern comparisons easier. No systematic differences were observed in the data based upon who prepared each specimen or which compound powder was deposited.

The binary compounds generally yielded the expected results with minimal effort in terms of post-measurement data processing.

### 7.2.2 Phase Identification Results

As was the case with the single-phase specimens, routine phase identification methods easily correctly identified the two phases present for each specimen prepared. Correspondingly, visually apparent differences in the micrographs had no observable consequence on the accuracy of the phase identification analyses performed.

### 7.2.3 Specimen Micrographs

Material effects (e.g., localized agglomerate deposits) were more noticeable in the micrograph images but these agglomerates may be partially related to the inexperience of all of the preparers working with the new 50-mesh version of the microsieves.

It is possible that the materials' tendencies were partially exacerbated by the users holding the microsieves with a shallower inclination angle to control powder flow characteristics. This version (i.e., 50 mesh) of the tool passed powder material in a fraction of the time required using the 60-mesh version. Following this thought process, to this point all of the users were used to the (slow) flow characteristics of the 60-mesh version of the tool. Perhaps the most compelling evidence of a "learning curve" effect using the 50-mesh microsieves can be found by comparing the appearance of agglomerates in Figures AII-2 through AII-10 with AII-12. The Binary F compound was deposited last by all of the participants. At the same time, the two compounds which comprised binary mixture F (burkeite and nitratine) exhibited the strongest agglomeration tendency in the single-phase specimens (see Figure AI-6 for burkeite and AI-8 for nitratine).

## 7.3 Five Saltcake Phase Compound Results

### 7.3.1 Diffraction Pattern Repeatability

On visual inspection, there were not obvious significant differences in the diffraction data traces.

### 7.3.2 Phase Identification Results

There are significant differences in the phase identification results from one pattern to the next.

In all nine specimens prepared, sodium nitrate and sodium oxalate were identified in the first round of the phase identification process when the entire database was searched. Subsequently in the second round, in eight of the nine specimens, burkeite was identified by searching the minerals subfile. Natrophosphate was identified in eight of nine data sets but required manual entry in half of the cases in order to be identified. Thermonatrite was significantly more problematic. In five of the nine specimens, thermonatrite could not be identified in the XRD data even when this phase was manually entered into the analysis software.

Several potential explanations could be proposed. First, phase segregation could happen while the powder mixture passed through the 50-mesh screen, thereby contributing to phase identification difficulties. In this scenario, user handling of the microsieve (as previously discussed regarding agglomeration tendency) may have enhanced or retarded the transmission of problematic particle geometries and their parent phases, resulting in lower average phase fraction (nominal 20%). This effect combined with the rapid data acquisition conditions used (compared to a normal acquisition speed ten times slower) would have diminished the overall statistical quality of the diffraction data making accurate identification more challenging. Furthermore, another factor that could have detrimentally contributed to the difficulty in identifying the phases present could be related to instrumentation errors. For the MiniFlex II, peak position errors were calibrated away rather than being corrected by methods of optical alignment. This instrument is a very basic model and by design, does not include mechanical alignment adjustments for the optical elements in the beam path. Accurate peak position data is acquired by a calibration process that shifts the scale of the data. This process, however, does not correct for defocussing effects (i.e., peak broadening). Defocussing causes peaks to become broader and, correspondingly, to have a lower relative peak height. Both of these effects (i.e., broadening and reduced peak intensity) make accurate peak identification more difficult under the best of circumstances. Further considering that the compound under evaluation is a multi-phase mixture, the probability of strong peaks from one phase obscuring the presence of weaker peaks from one or more other phases becomes increasingly likely. One result of this sequence of events is that major phases are easily identified while multiple other diffraction peaks remain unidentified.

### 7.3.3 Specimen Micrographs

As likely as the case just presented might appear to be, another compelling explanation also exists. In all five cases where thermonatrite could not be identified, review of the associated specimen micrographs revealed that the deposit surface had some form of mechanical disturbance after the powder was deposited. In four of the five cases mentioned, the disruptions were during the mask removal step (i.e., the hold down tool slipped, marring the deposit in the process). In the other case, the mask material (tape) curled upon release from the silicon surface and dragged a corner across the deposit surface leaving a thin scratch through the deposit surface.

### 7.3.4 Phase Identification Sensitivity to Preparation Errors

The damaged specimen deposit explanation is deemed the more reliable argument between the above two discussions presented. This is principally due to the fact that all five phases were accurately identified in some of the data sets acquired and in one case this was achieved in the second round of the identifications (i.e., successful phase identification in two mouse clicks). The microsieve-calibration-rapid acquisition explanation should have been equally or at least nearly equally detrimental to all nine data sets.

It is likely that the following factors contributed to thermonatrite and natrophosphate not being identified in all of the prepared specimens:

1. The thermonatrite simulant was not phase pure, meaning that the five-phase mixture included less than 20% thermonatrite by weight,

2. The mechanical disturbances not only decreased the area of the powder deposit but likely also created a beam-obstructing high spot that would have interfered with detection of diffraction peaks below 17 deg 2Theta.

3. The petroleum jelly adherent used included two crystalline peaks, and the lower angle peak (see Figure 3) significantly overlapped an important thermonatrite peak,

4. The patterns were acquired 10x faster than is standard for tank waste specimens. The over 100 specimens in this study and a low power/low resolution instrument with a monochromator combined with a faster acquisition time resulted in a higher statistical perturbation than is standard in tank waste data sets. The higher noise level made identification of overlapping diffraction peaks that much more difficult to associate with unique phases.

5. Natrophosphate was not identified in one of the nine samples but it also had to be manually inserted in six of nine data sets in order to be identified. It is suspected that in spite of being ground several times, this particular salt retained larger particle sizes than the other salts (reduced particle statistics, thereby reducing data quality and, therefore, detectability). This problem was further exacerbated by the mechanical disturbances mentioned previously.

Analytically speaking, the phases most easily identified had the majority of their most significant peaks above 30 degrees 2theta. All three saltcake phases with a significant number of peaks below 20 deg 2theta were increasingly difficult to identify in the data, with thermonatrite and natrophosphate being consistently the most troublesome.

## 8. LESSONS LEARNED

The petroleum jelly adherent enabled reliable deposition of randomly oriented powder particles. At the same time several additional details were revealed relating to this material. First, this material has an optimal temperature range (~80 to 87 ºF). Since it melts at 90 ºF, temperatures close to this or above result in the powder sinking into the petroleum jelly. This enhances the diffraction of the petroleum jelly while also diminishing the signature of the analyte powder. Both of these results are counter to the purpose of accurately identifying phase content in the analyte powder. At the same time, at temperatures significantly cooler, the petroleum jelly becomes less compliant and less adherent with deposits being increasingly sparsely populated as the temperature drops.

The second consequence of using the petroleum jelly related to the transformation of the original deposited powder into a contiguous crust. In general, it is not desired to have a deposited material change. However, in the specific case of mounting radioactive powder deposits, this turned out to be a real advantage in terms of diminishing the potential for spreading contamination while the specimen is outside the hood environment (e.g., while being transported to/from instruments and during evaluation processes on instruments). This transformation process did entirely obscure the possibility of evaluating individual implanted particle orientations thus limiting the extent to which the specimen preparation method could be scrutinized analytically.

A related but likely detrimental process associated with the powder-to-crust transformation process was the weight loss observations made. No evidence of distortion of diffraction data was identified. However, it is plausible that the diffraction response of trace phases could potentially be diminished below detection limits should the transformation process diminish or otherwise mask their scattering characteristics.

The third consequential effect of using the petroleum jelly adherent is the phase specific interference that occurs between thermonatrite, trona, and the petroleum jelly peak near 2theta = 21.4 degrees. Diffraction peak overlap is difficult to avoid in the analysis of multi-phase unknown compounds. Adding the petroleum jelly crystalline peaks to the mix is other than beneficial.

The microsieve with its variants (e.g., 40-, 50-, and 60-mesh types) proved to be highly successful in terms of controlling powder deposition. At the same time the “art” of the method (i.e., handling practice or “hand feel” for ideal powder deposition) is not definitively scripted at this point. The interaction between tool, analyte material, and wielder of the tool may require “time with the tool,” the extent of which may need to be determined on an individual basis.

Imaging practice with regard to documenting specimen mounts was proven to be exceptionally valuable in interpreting the diffraction data. Given the deposit transformation process that was documented in this report, it may be better practice to allow the transformation process to go to completion. This would ensure that the diffraction data collected corresponded to the micrograph features being interpreted in conjunction with the diffraction data. In other words, the deposited powder form has proven to be transient, therefore it is proposed that documenting the steady-state (final) form would be more appropriate. A buried detail in this is that the diffraction scans were typically run with six specimens being processed in succession (approximately 8 hours of measurement time). Even in the cooler parts of the year, this was more than enough time for all of the specimens to fully transform.

# 9. CONCLUSIONS

The specimen preparation techniques described in this method development study built upon an existing XRD specimen preparation method and added aspects that tailored the final method to the preparation of saltcake samples and, more generally, to solid tank waste materials to be characterized for solid phase content. The thin powder deposit method developed was found to be superior to the packed powder cavity method in terms of generating high quality reliable data and also by substantially reducing the volume of material used to generate XRD specimens. Using a smaller volume of material clearly reduces the exposure of the individual preparing radioactive specimen mounts and also limits the potential spreadability of contamination associated with the transport and characterization activities performed outside the fume hood environment.

At the same time, this method was found to have vulnerabilities involving mechanical disturbances to the deposited material. That being said, mechanical disturbances to the deposit are very much easier to visually identify than the preferred orientation problem encountered with packed powder mounts. While it is possible to minimize the effect of a single mar spot (e.g., orient the mar parallel to the diameter of the X-ray beam and choose not to spin the specimen during data acquisition), the recommended practice is to prepare another specimen mount. Since this specimen preparation method takes less than one minute, the benefits of preparing another (with the existing tools and materials remaining in the mortar) are deemed worth the additional potential exposure given all of the other exposure minimizing practices in place.

## 10. ALARA COLLABORATION

Many tools and practices were developed (and described herein) in support of XRD specimen preparation method development. However, the designs were conceived in collaboration with at least 20 individuals throughout the 222-S complex, and the final designs would not have been as developed without the participation of many others. In this section of the report, specific ALARA enhancements, achieved in part by collaboration with other staff members from other parts of the 222-S laboratory, are discussed. Staff and management from Special Analytical Services and Process Chemistry collaborated with the staff and management of the organizations cited below to enhance the ALARA characteristics of the tools and components cited.

[Collaboration with Prod Ops 222-S & Tank Farms Surveillance]

**Acrylic Shelf** – This hood storage component allows “in-process” sample materials to be moved out of the active work space to a position that provides greater distance and shielding potential for individuals working in the hood space. In addition, it also provides floor space for other dose emitting items (e.g., solid waste and prepared specimen storage – see Figures AIV-1 and AIV-2 in Appendix IV).

**Acrylic Mortar Support Plate with Disposable Rubber Insert** – The acrylic base is an original design constructed from acrylic sheet stock (see Appendix IV, Figure AIV-2). The rubber insert was made from the lower half of the same commercially produced part that is used as a shield for “in-process” samples (see below). This assembly allows an “in-use” mortar to be stabilized keeping the user’s hands at a greater distance. The non-porous nature of the components allows for easy cleaning and, in the case of the rubber insert, this part can be disposed of in instances where spillage from the mortar contaminates it. In addition, this device also provides a degree of shielding when “in-process” materials are stored on the associated acrylic shelf.

**Microsieve External Tungsten Shielding** – This product can easily be added to the microsieve design on an "as-needed" basis providing beta/gamma emission shielding where close distance is a very limited ALARA parameter (see bottom caption of Figure 9).

[Collaboration with Production Ops 222-S Radiological Controls]

**Rubber Cone Shield** – This is a commercially produced component that was re-purposed as disposable shielding. This vented part allows "in process" samples to continue drying while also providing shielding to users of the hood environment. In addition, this part can easily be wiped clean if contaminated or simply disposed of due to the small size and low cost (see Appendix IV, Figure AIV-2).

**Microsieve Waste Container** – The polyvinyl chloride (PVC) container allows used microsieves to be disposed of in a compact and shielded container that easily accommodates the natural dimensions of the tool (see Appendix IV, Figure AIV-1).

[Collaboration with 222-S Facility Operations; Laboratory Facility-Maintenance Mechanical]

**Extension Pestle** (prototype and revision) - The shaped rubber handle provides both mechanical leverage while also providing shielding from sample emissions from the analyte being ground. The original single piece handle provides 6 times the distance of the original commercially produced agate pestle, with the revised version providing a mechanical separation point via a repurposed plumbing fitting. The addition of the separation point allows the contaminated portion of the tool to be wiped, then separated and placed in an ultrasonic bath without the user having to support the top-heavy tool (minimizing potential dose exposure to the user - see Appendix IV, Figure AIV-2).

## 11. FUTURE METHOD DEVELOPMENT OPPORTUNITIES

Given the vulnerabilities of the petroleum jelly adherent, an alternate adherent with equivalent implantation characteristics but fewer vulnerabilities is currently being considered. Beyond the limited scope of XRD specimen mounts, it is desired to develop a specimen-mounting methodology that would be equally suitable for PLM and scanning electron microscopy evaluation as well. The petroleum jelly adherent is not well suited to either of these solid phase characterization methods. Were such a unified specimen preparation method to be successfully developed, there would be ALARA benefit, as well as greater potential for unified interpretation of analyte material phase composition.

The ALARA benefit should be obvious in that multiple individuals are currently exposed to radioactive materials during specimen preparation activities.

A unified method specimen mount would allow the same single mount to be evaluated on three different instruments. The analytical benefit in part relates to the elimination of sub-sampling mysteries. For example, when a multi-phase mixture is sub-sampled it is always possible for each sub-sample to be inhomogeneous. Even worse, it is also possible for each subsampling process to completely remove or mask one or more low fraction components of the sample.

Subsequent resolution of different results produced with different instrument and specimen preparation methods can be problematic.

This method development study also revealed some of the measurement limitations of the existing 222-S Laboratory instrumentation. More specifically, the adherent interferences could easily be mitigated if not completely eliminated by using a diffraction instrument capable of performing grazing incidence diffraction measurements. In this measurement mode, the X-ray beam has a shallow incidence angle that is held constant during the entire acquisition process. This x-ray beam geometry geometrically limits the penetration of the X-ray beam, thereby emphasizing the scattering response of the deposited powder. Furthermore, most research grade diffractometer models are more powerful, resulting in higher quality data more quickly, and they include the capability to perform other measurement types such as transmission measurements where the X-ray beam passes through a permanently sealed glass capillary (or equivalent vessel). Clearly there is science benefit, but ALARA benefits would also be derived both in that the higher power instrument is capable of evaluating permanently sealed specimens and the data quality per unit of dose received by the preparer would unambiguously be improved.

## 12. QUALITY CONTROL

All testing carried out under this test plan followed ATS-MP-1032*, 222-S Laboratory Quality Assurance Project Plan*. All relevant details regarding the solid phase characterization instruments, scientists, project coordinator, logbooks, instrument procedures, and data acquisition dates are recorded on A-6006-872, "Solid Phase Characterization Analysis Tracking" form, which is saved in the project file. Quality control practices for each of the solid phase characterization methods used are described in the associated instrument procedures.

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# APPENDIX I: SINGLE-PHASE SALT SPECIMENS

Method I and Method II (both 60-mesh and 50-mesh variants) were used to prepare single salt specimens. Micrographs of the individual specimen preparations were sometimes taken within minutes of powder deposition. In other instances, the XRD measurements were acquired first and the micrograph images were taken after the specimens were removed from the MiniFlex II (i.e., many hours after the specimens were originally prepared). Early in this method development study it was not realized that the image quality would be impacted. As part of this method development study, an unexpected morphological transformation was revealed. The original powder deposits interacted with the petroleum jelly adherent in a manner that preserved the original phase content but fused the deposited powder particles into a contiguous polycrystalline crust. In the micrographs, untransformed powder deposits have a “snow-like” (bright white), appearance while the transformed specimens have a translucent “ice-like” (darker), appearance. Most of the composite images (i.e., Figures AI-2, AI-4, AI-6, AI-8, and AI-10) include both types (i.e., adhered powder & transformed crust).

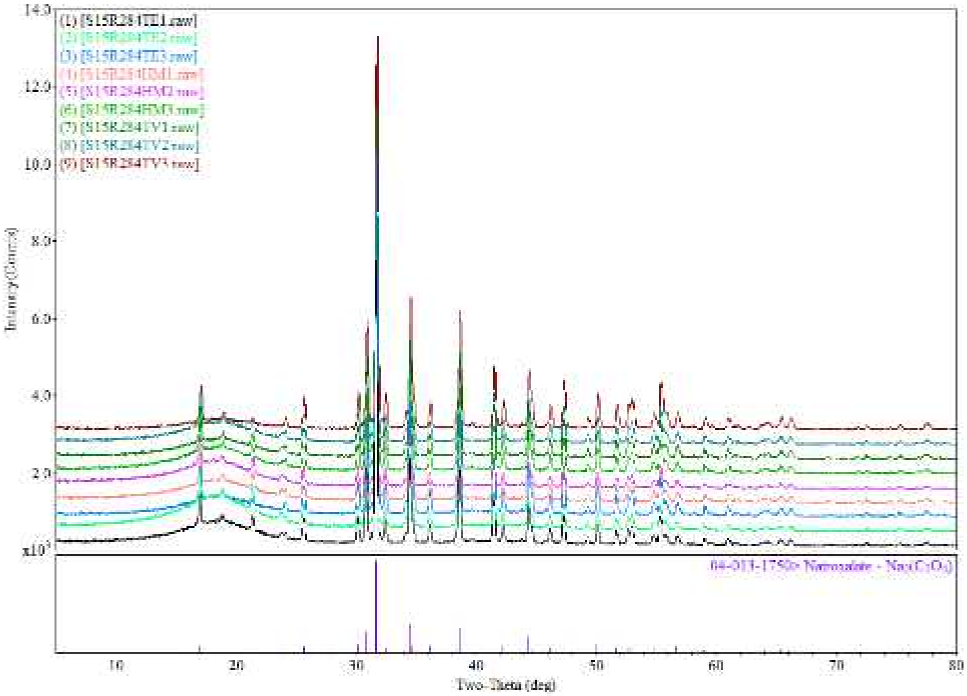


Figure AI-1. Natroxalate XRD data and phase content.

Figure AI-2. Natroxalate specimen preparations.

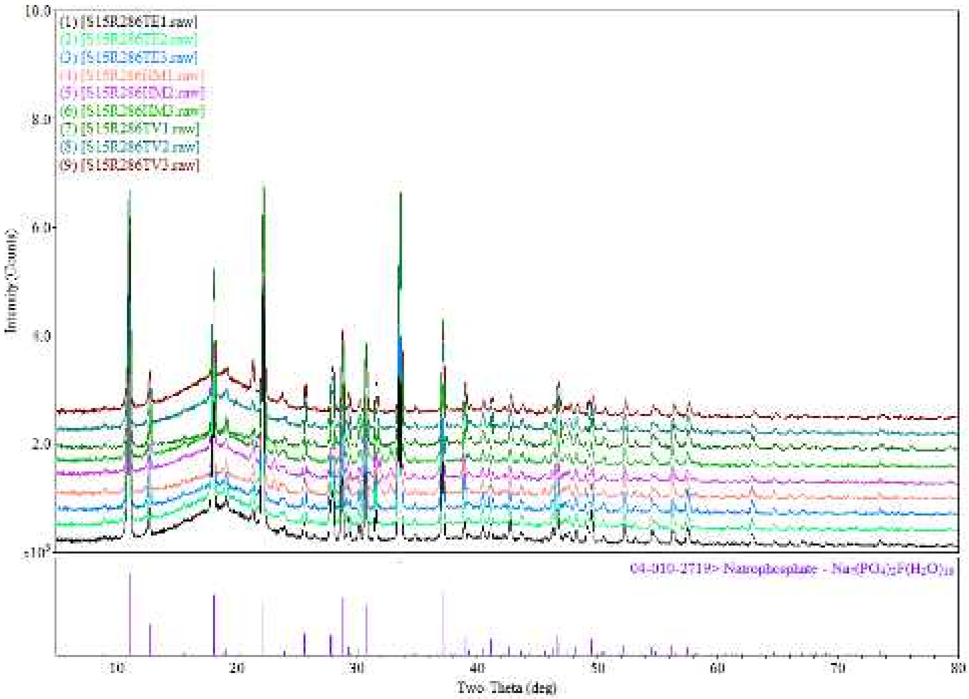


Figure AI-3. Natrophosphate XRD data and phase content.

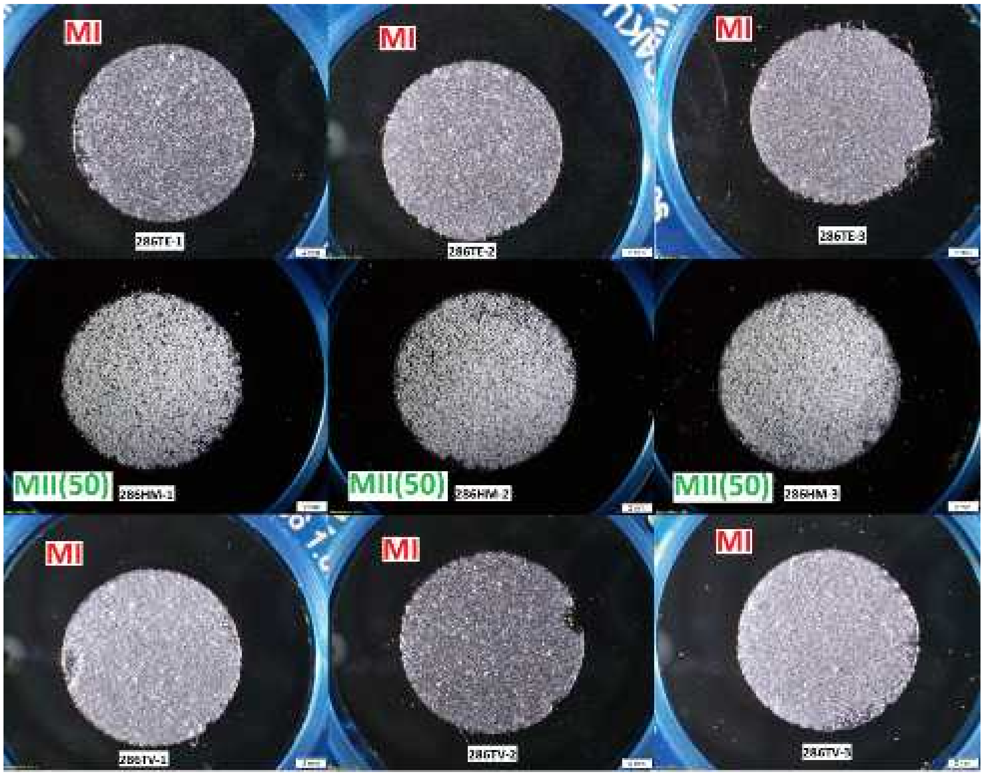


Figure AI-4. Natrophosphate specimen preparations.

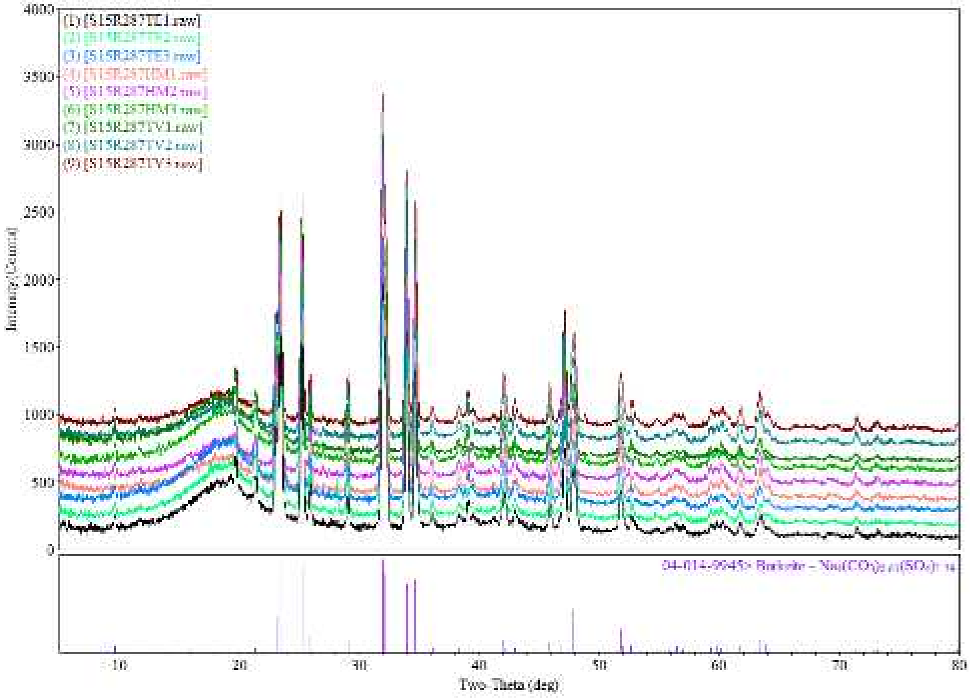


Figure AI-5. Burkeite XRD data and phase content.

Figure AI-6. Burkeite specimen preparations.

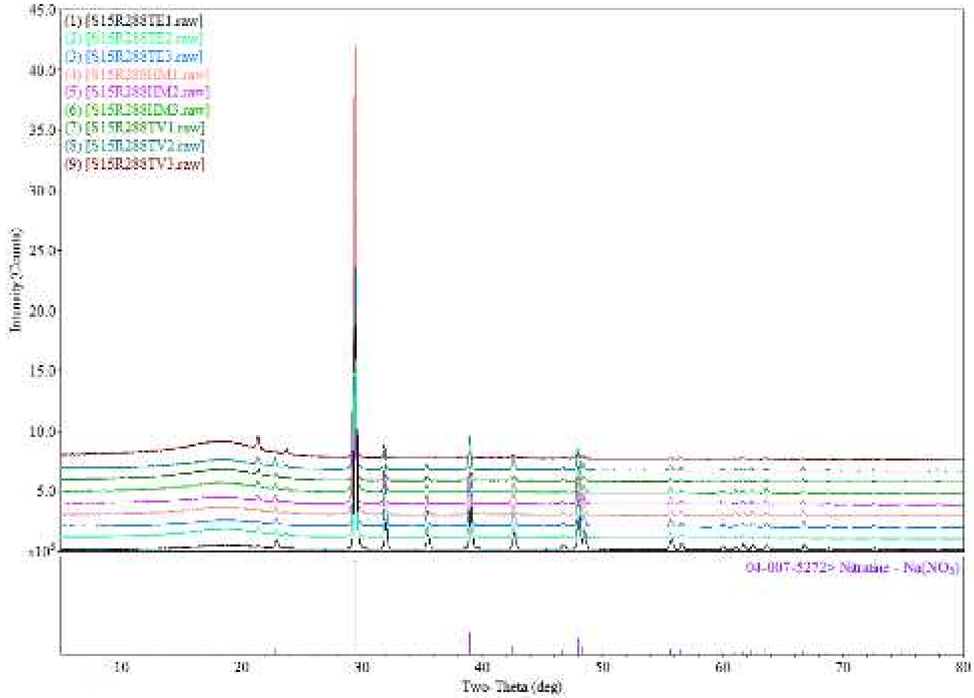


Figure AI-7. Nitratine XRD data and phase content.

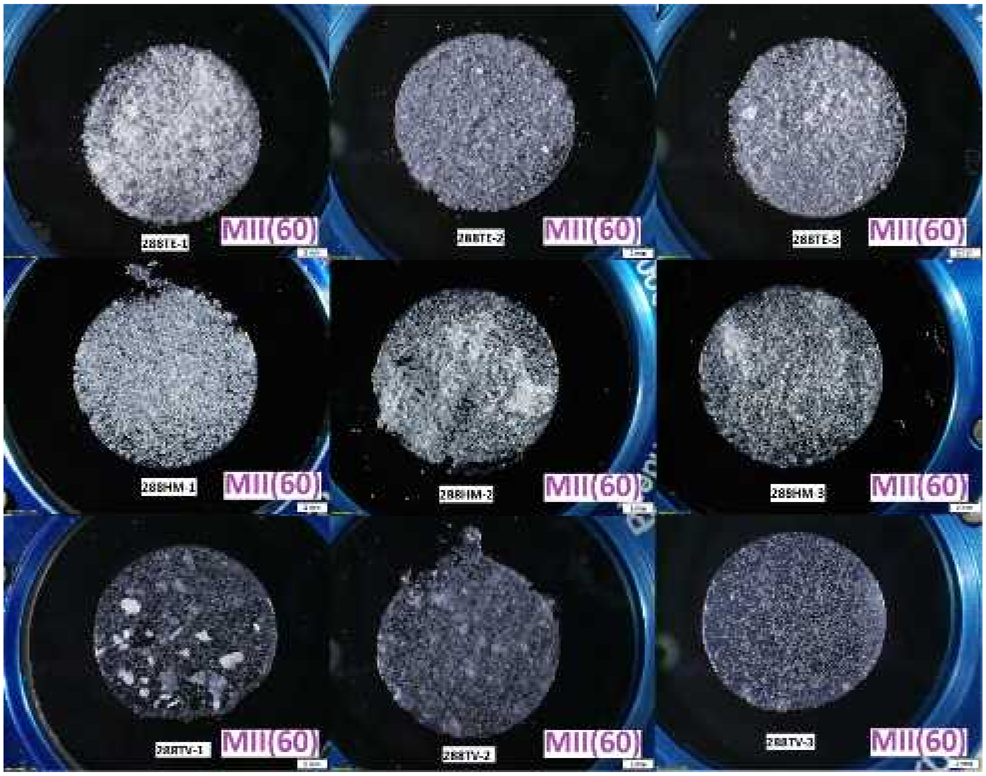


Figure AI-8. Nitratine specimen preparations.

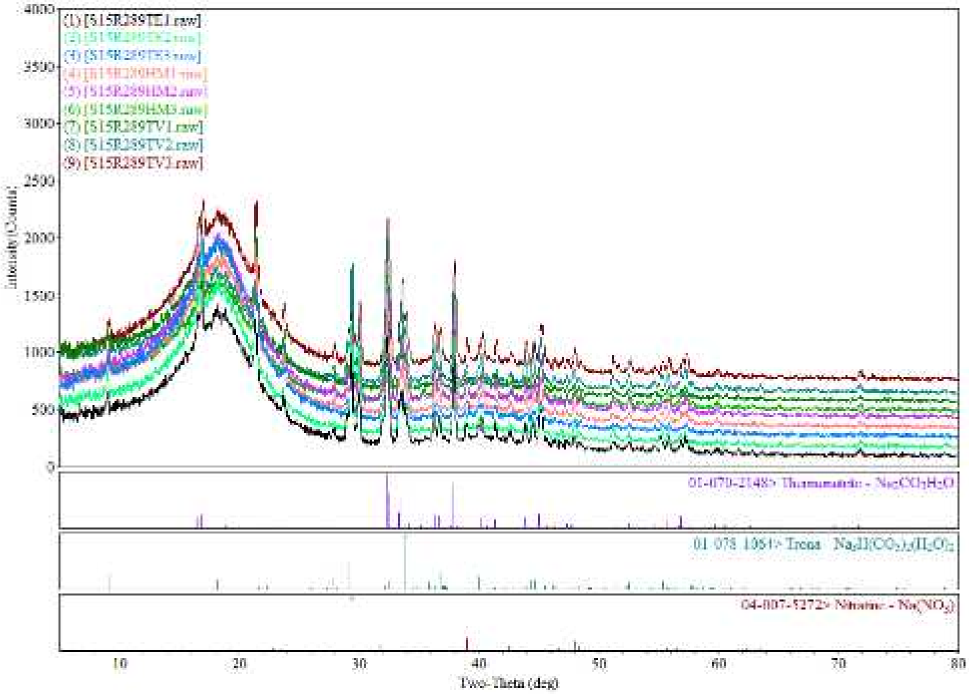


**Figure AI-9. Thermonatrite, trona, and nitratine. The latter two phases indicate the thermonatrite synthesis process was unsuccessful.**

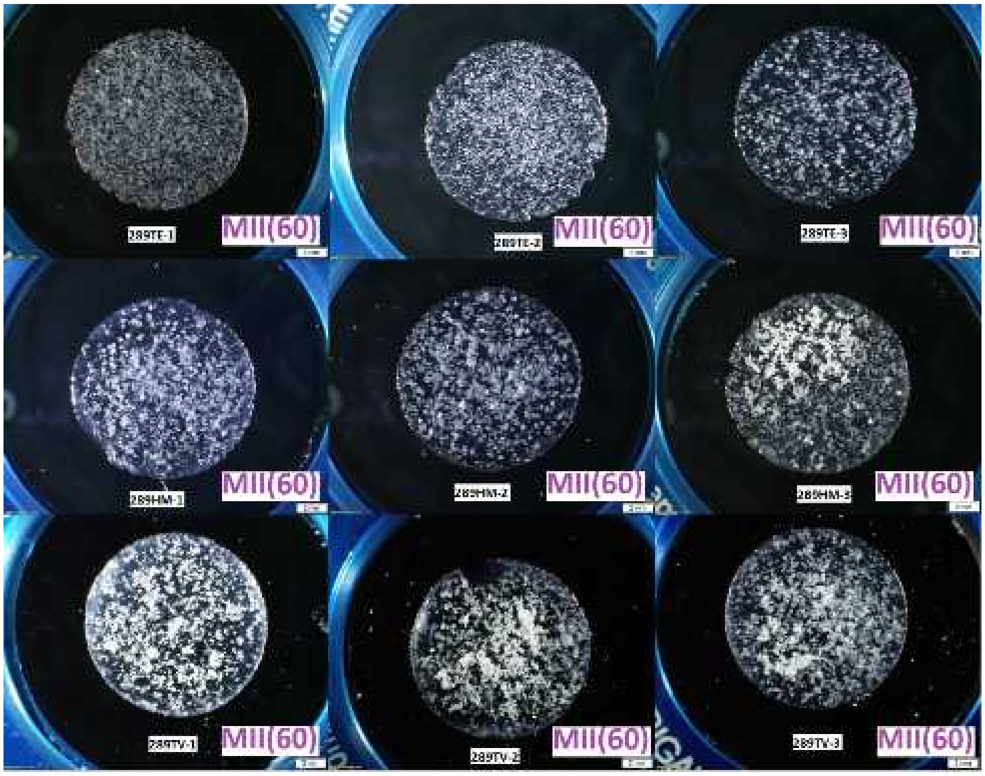


**Figure AI-10. Thermonatrite specimen preparations.**

# APPENDIX II: BINARY PHASE SALT SPECIMENS

As a result of experiences developed with the single saltcake portion of this project, Method II (with the 50-mesh screen) was exclusively used in the preparation of specimens for binary saltcake phase compounds. The micrograph of each specimen was taken prior to XRD measurements being performed with rare exceptions (individually noted). This was done in an attempt to capture specimen deposit features prior to transformation into a contiguous crust. The earliest of these specimens were prepared in mid-April, meaning that the ambient temperature in the laboratory was higher. At the same time it was informally noticed that the crust transformation process that had been taking up to a few hours was now only taking a few minutes to complete in the warmer months. In the composite images, the overwhelming majority of the images have a snowy-white appearance with few exhibiting mars or other non-uniformities.

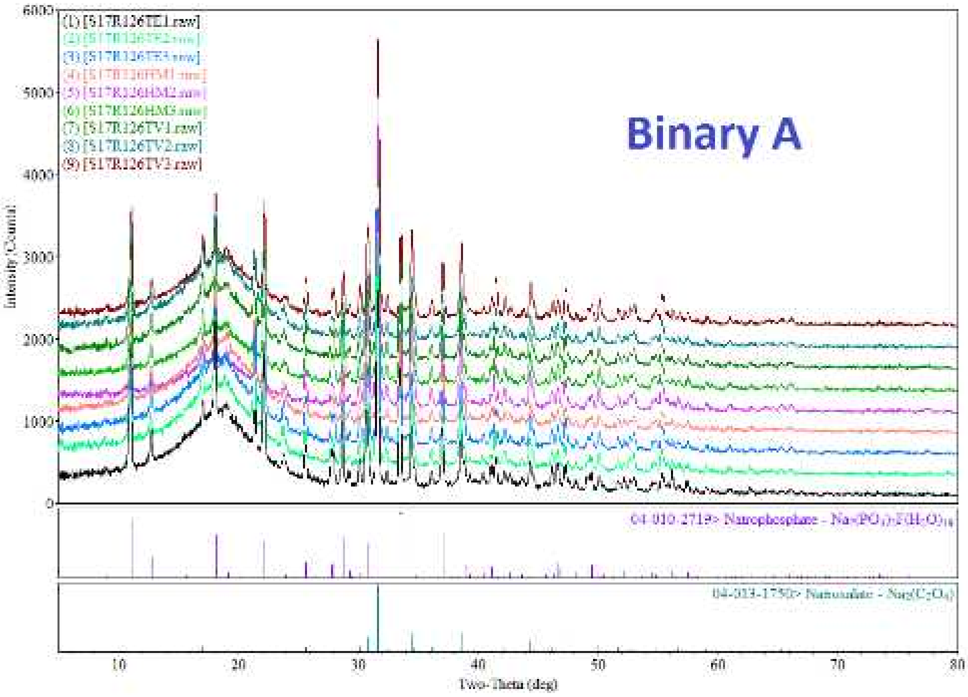


Figure AII-1. (50:50) Binary mixture of natroxalate and natrophosphate (Binary A).

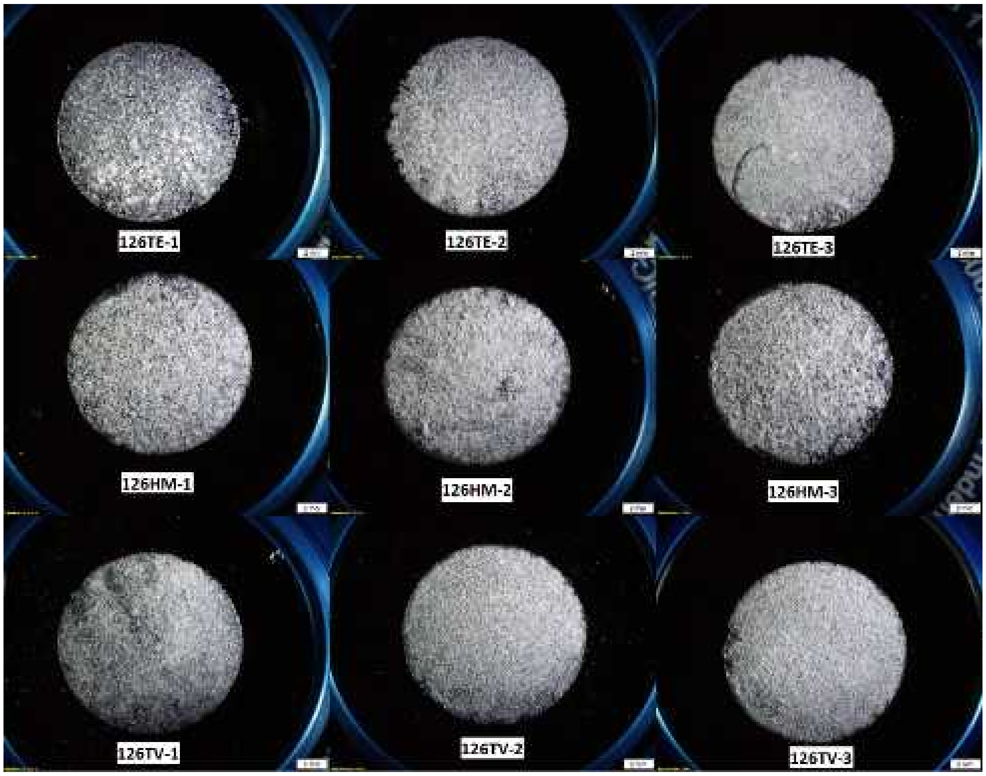


Figure AII-2. Binary A specimen preparations using Method II (50).

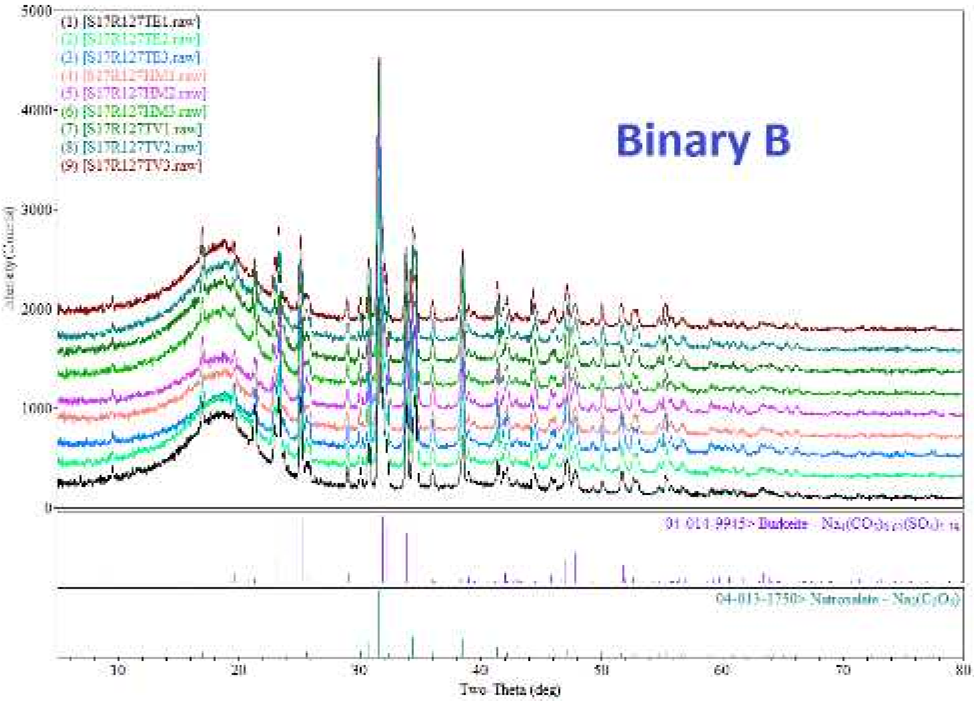


Figure AII-3. (50:50) Binary mixture of burkeite and natroxalate (Binary B).

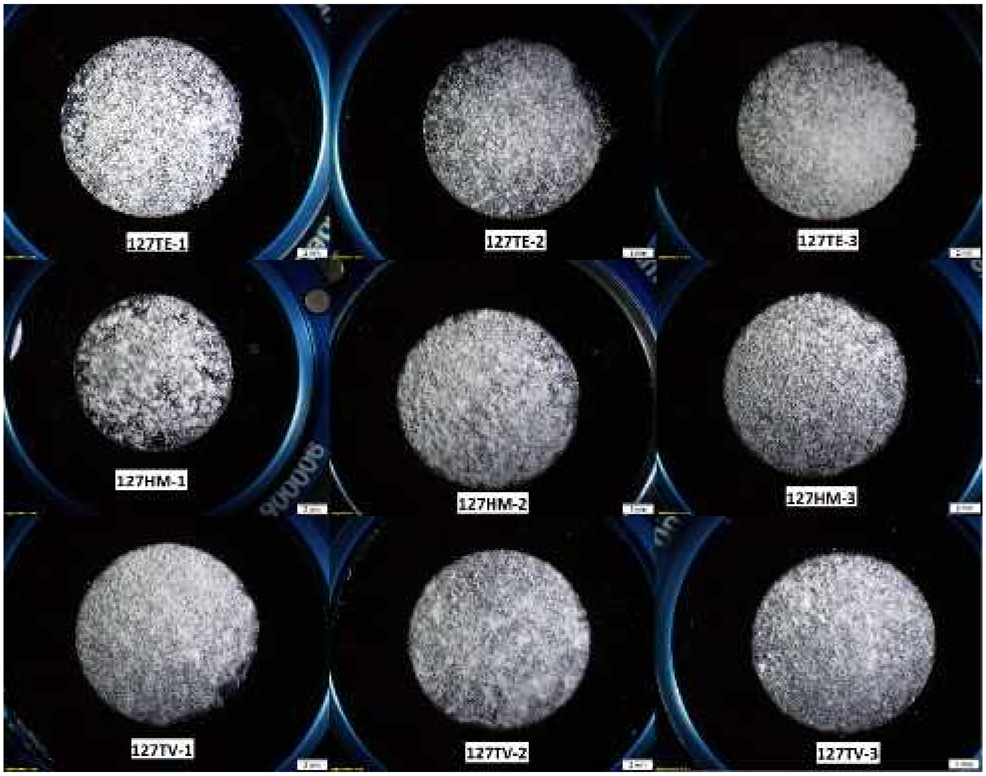


Figure AII-4. Binary B specimen preparations using Method II (50).

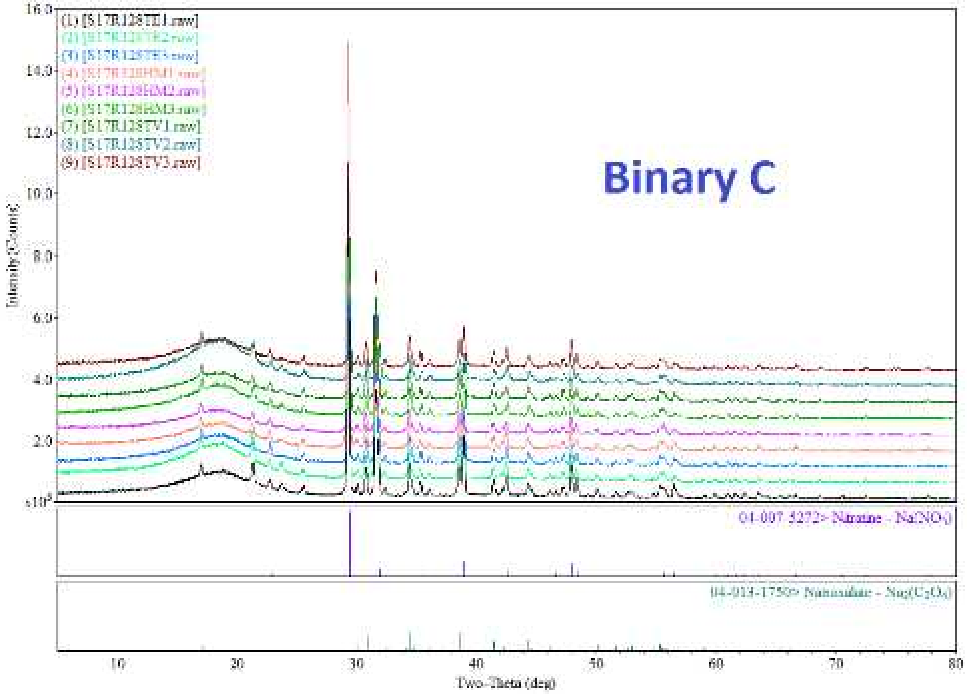


**Figure AII-5. (50:50) Binary mixture of nitratine and natroxalate (Binary C).**

**Figure AII-6. Binary C specimen preparations using Method II (50).**

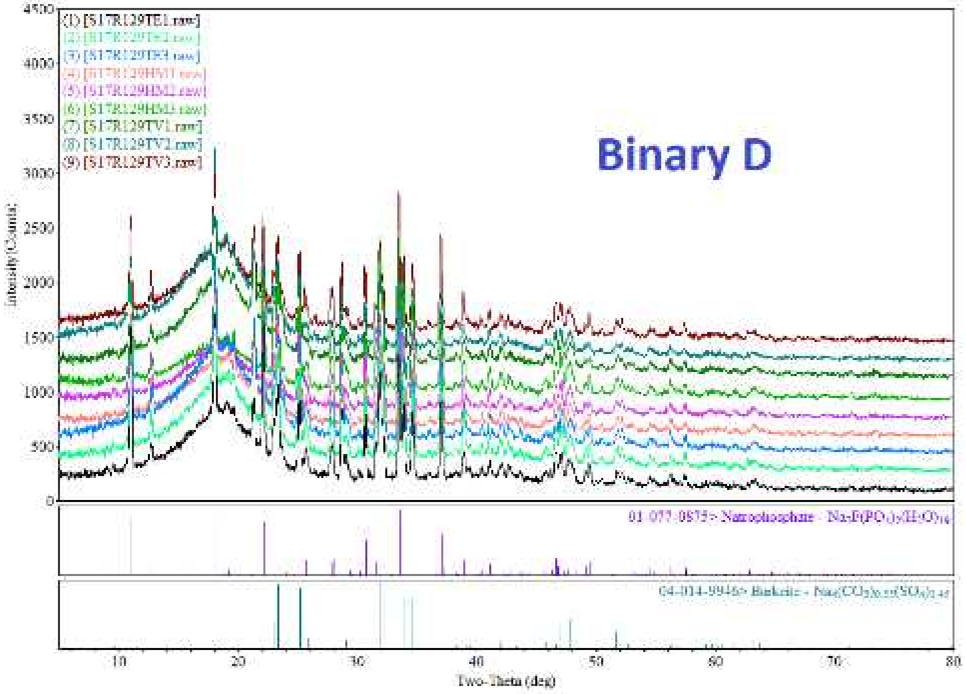


Figure AII-7. (50:50) Binary mixture of natrophosphate and burkeite (Binary D).

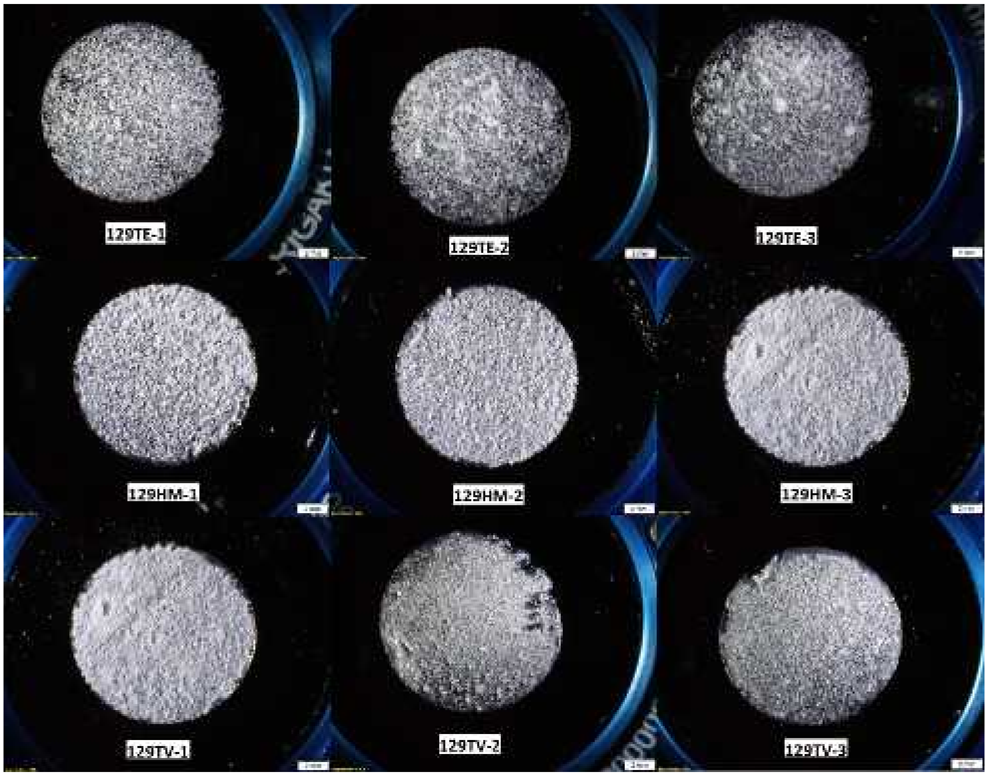


Figure AII-8. Binary D specimen preparations using Method II (50).

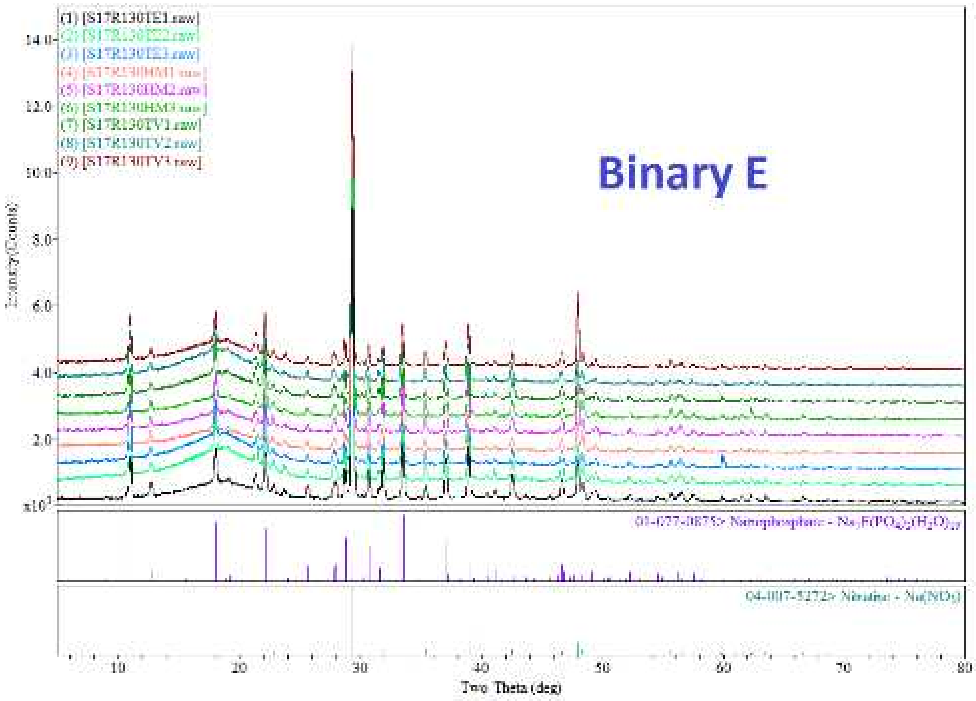


Figure AII-9. (50:50) Binary mixture of natrophosphate and nitratine (Binary E).

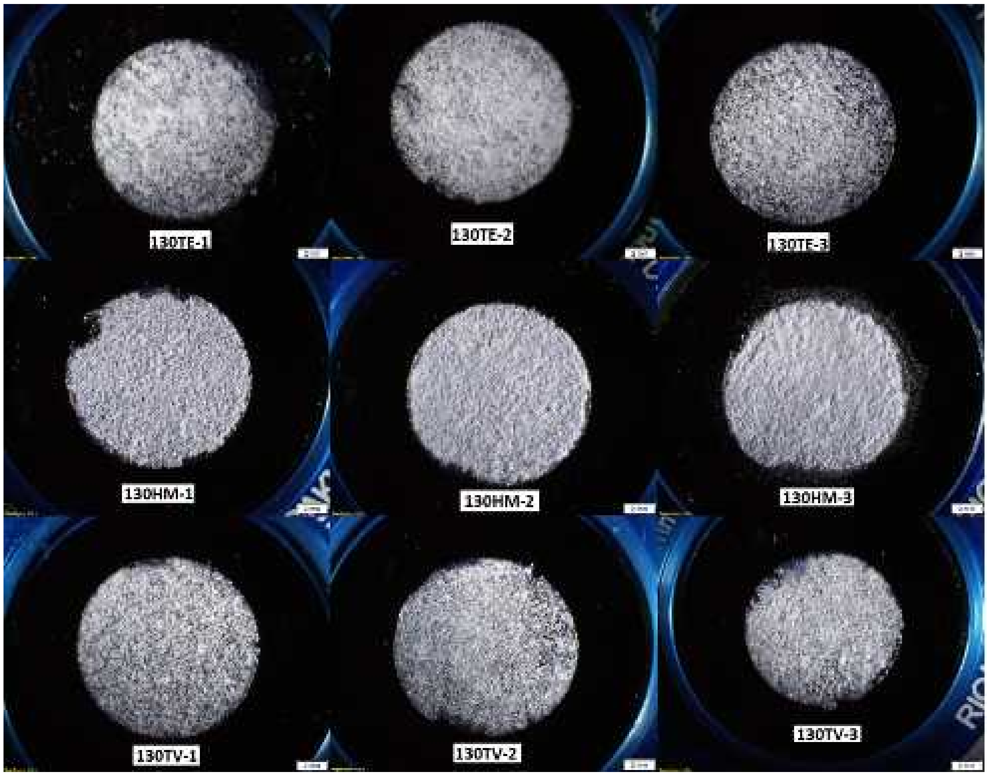


Figure AII-10. Binary E specimen preparations using Method II (50).

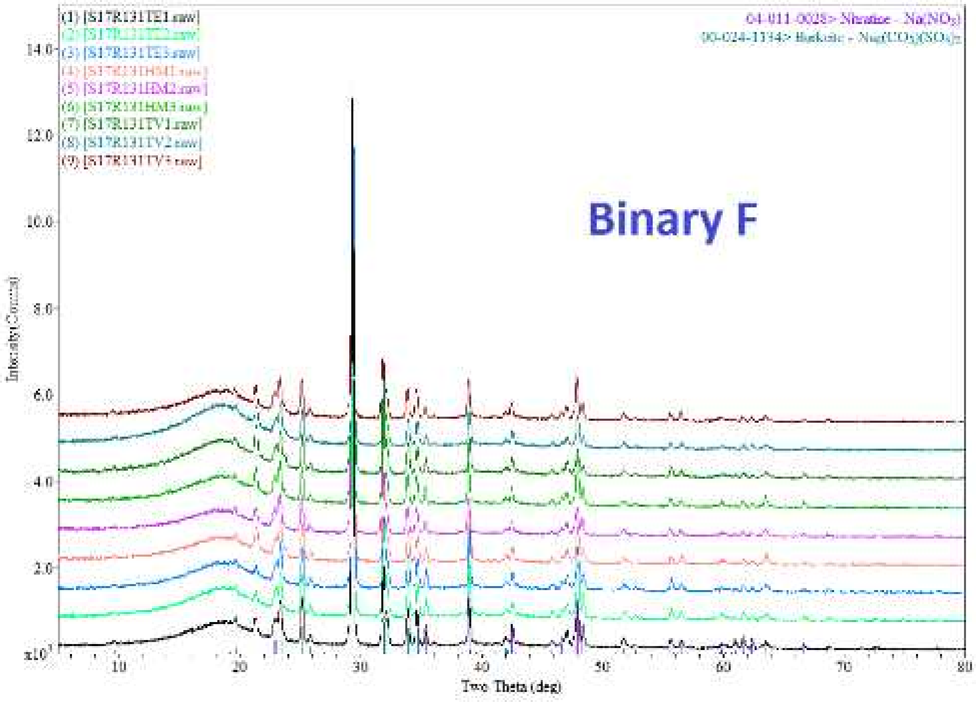


**Figure AII-11. (50:50) Binary mixture of nitratine and burkeite (Binary F).**

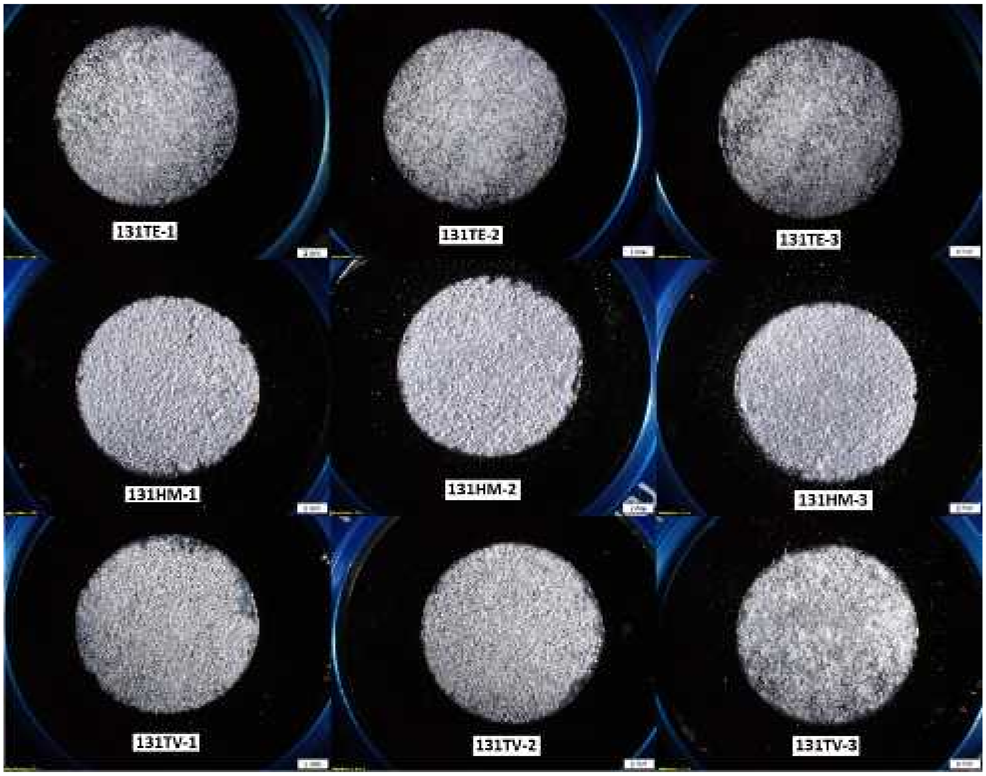


**Figure AII-12. Binary F specimen preparations using Method II (50).**

# APPENDIX III: FIVE-PHASE SALT SPECIMENS

As was the case with the binary mixtures, Method II (with the 50-mesh screen) was exclusively used in the preparation of specimens for the five saltcake phase compound. The micrograph of each specimen was taken prior to XRD measurements being performed.

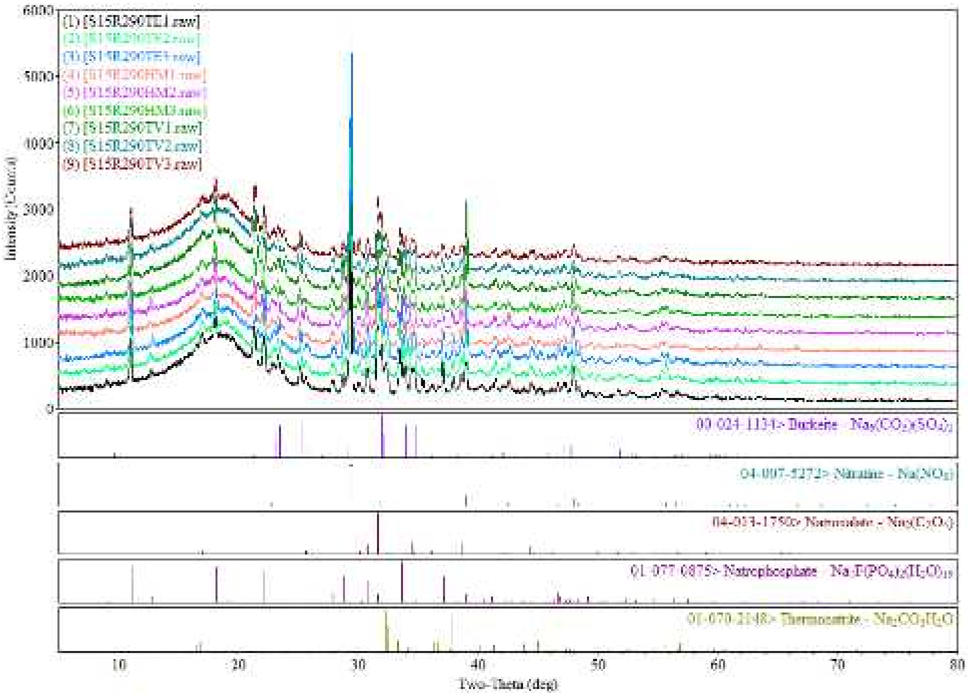


**Figure AIII-1. 20% by weight mixture of all five saltcake phases.**

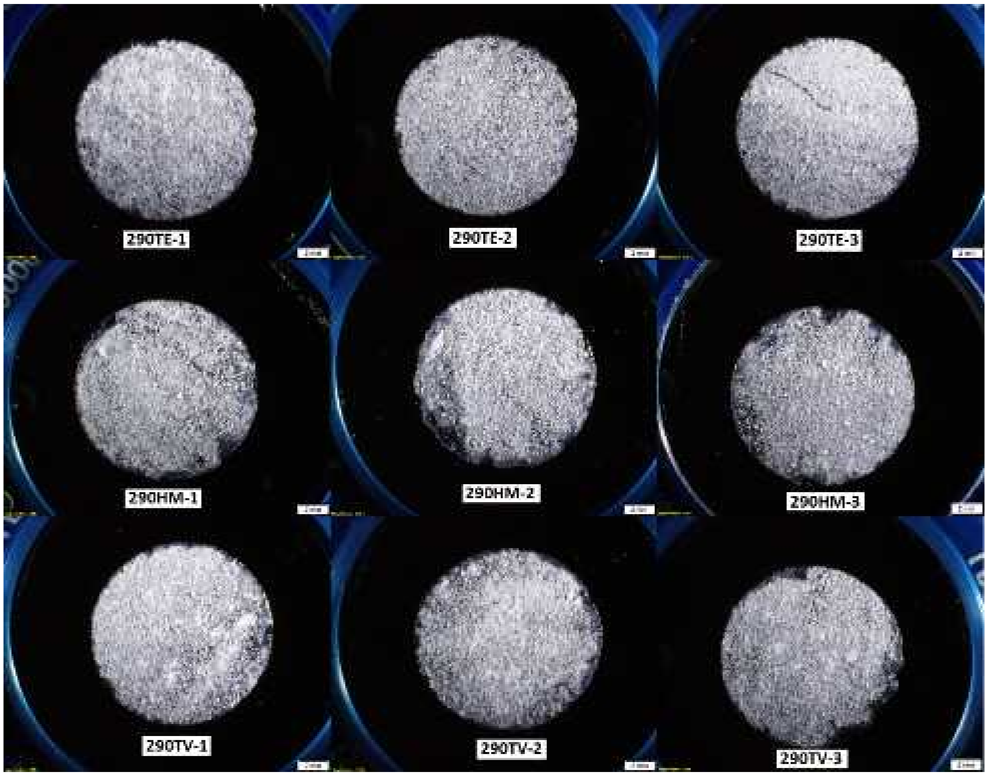


**Figure AIII-2. Five-phase specimen preparations using Method II (50).**

# APPENDIX IV: CONTAMINATION AREA FUME HOOD CONFIGURATION FOR XRD SPECIMEN PREPARATION AND ASSOCIATED TOOLS

## XRD Instrument and Specimen Preparation History in the 222-S Laboratory

From first installation until June 2015, the Rigaku MiniFlex II was in Hood 4 in room 1GA. In June 2015, it was moved to a countertop location next to hood 1 in room 1GA, and the majority of all specimen preparation activities were moved from hoods in room 1D to hood 1 in Room 1GA (vacuum filtration of wet or moist sample materials continues to be performed in hood 3, room 1D, with the dry filter cake solids being transported to hood 1, room 1GA, for further specimen preparation processing).

Formerly, hood 1 was associated with a moisture analyzer instrument and was extensively reconfigured in support of XRD specimen preparation activities. This appendix describes the reconfiguration of hood 1 and the associated development of tools in support of XRD specimen preparation and post-analysis disposal of specimen mounts. Figure AIV-1 shows all of the major components of hood 1 in room 1GA after it was reconfigured principally for the preparation of XRD specimens.

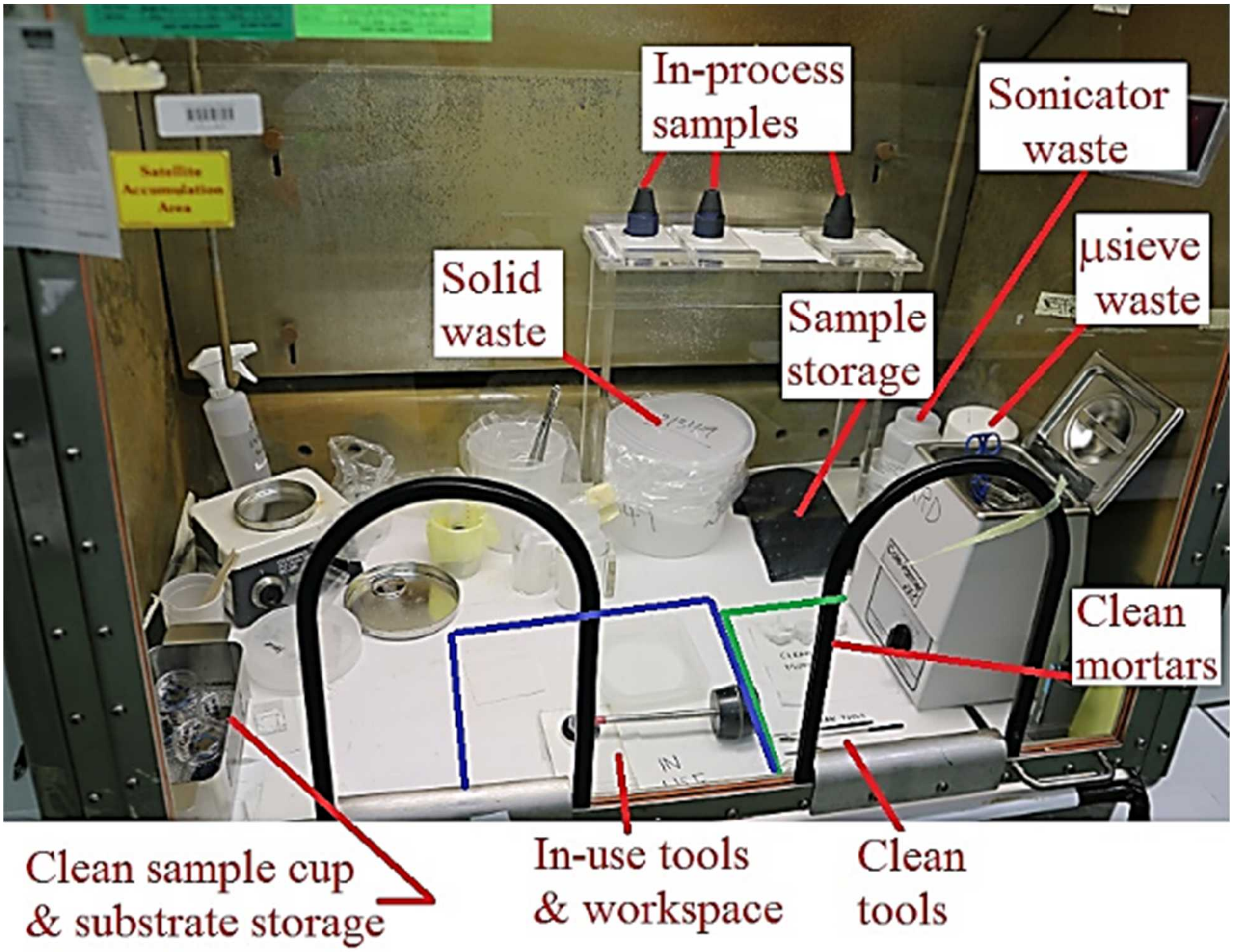


**Figure AIV-1. XRD specimen preparation apparatus are shown in this image.**

The design of the hood layout is intended to be ergonomically advantageous to individuals preparing XRD specimens. In general, solid and liquid waste containers are positioned on the right rear corner of the hood to provide as much of a distance component between anyone entering the hood and the waste. Correspondingly, the layout of the left side of the hood should typically be free of radioactive materials (samples or waste), thereby providing a lower dose portion of the hood. The work space is intended to be located toward the front of the hood between the arm ports, providing convenient access to tools and accessories otherwise located around the periphery of the work space. In-use articles are intended to be moved from the clean space on the periphery into the work space and remain there until they have been fully cleaned and dried.

Specialized tools were developed in order to improve ALARA aspects of specimen preparation as indicated in Figure AIV-2. The 1-inch diameter agate mortar and matching 1-inch long agate pestle are commercially available products that were selected in part due to their limited size and non-porous nature but were subsequently modified in order to provide greater distance between the individual performing grinding operations and the radioactive media being ground.

**Figure AIV-2. Specialized tools developed for XRD specimen preparation are shown in this composite image. (Upper left to lower left): Prototype and follow-on design extension pestles; (Upper center to lower center): Mortar support plate components; (Right): Acrylic shelf with upper drying rack, lower area for solid waste and specimen storage under black rubber sheet shielding.**

In particular, the original pestle prototype design was a one-piece tool with the agate potted in place using potting epoxy at one end of a 6-inch long stainless steel tube section. The handle was constructed of two size 12 rubber stoppers bonded together after one was supplied with a center through hole (diameter sized slightly smaller than the outer diameter of the stainless steel tube). The tube end was fully inserted into the rubber handle. The design of the rubber handle provides a mechanical advantage in addition to providing the user with 2 inches of rubber shielding between the sample material being ground and the user's fingers and hand. The tapered shape of the handle allows the user to grip the top half of the handle, minimizing the component of direct shine dose received by the user while performing grinding of sample materials.

The acrylic plates were designed to provide stable support for the mortar while grinding is being performed. This allows the mortar to be pressed in the rubber ring prior to the introduction of sample material and holds the mortar tightly while grinding is being performed without requiring hand support in the near vicinity of the mortar. Typically a tabbed piece of packing tape is placed across the bottom of the hole in the acrylic plate to ensure that the rubber ring (made from a modified Gooch crucible holder) and mortar do not slip out when the plate is lifted. Since the acrylic plate has a raised edge in the front, this is a convenient location for writing the sample designation for the material being processed (typically done using a black marker with alcohol-soluble ink). The length of the plate provides distance between the ground radioactive materials and the hand of the person moving the ground sample materials to or from the acrylic shelf. Meanwhile the thickness dimension of the plate plus rubber ring plus mortar wall thickness act as shielding between the ground materials and the hand holding the plate.

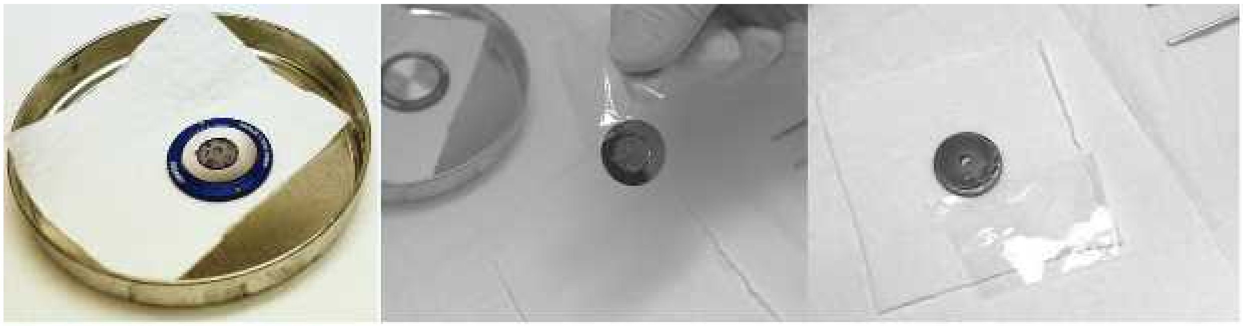
In the course of processing sample materials, the vented rubber cap (i.e., Gooch crucible holder) is usually placed over the ground material while evaporation of the liquid grinding media (e.g., ethanol, isopropanol, and acetone) is occurring. The cap provides shielding for individuals working in the hood while also providing protection of the ground sample materials from potential airborne particulates comingling with sample materials. This is an increasingly important consideration as the number of samples "in process" increases and/or in situations where the sample materials are more likely to self-mobilize as the drying process proceeds. After sample preparation has been completed, the design of the support plate allows for easy disassembly and cleaning. The rubber support ring is considered a consumable and is typically discarded if radioactive contamination is detected.

The acrylic shelf has several advantageous design features. First, during the design phase acrylic was selected as the construction material because it is non-porous (making it relatively easy to clean and decontaminate), and since it is transparent it does not make dark zones inside the hood.

Second, the eye-level shelf was intentionally designed to elevate the samples "in process." The acrylic shelf thickness in combination with the plate components described above provide a degree of shielding from alpha and beta radiation sources thereby lowering the dose received by anyone working in the hood. Third, moving the "in process" materials out of the active work space allows new sample materials to be processed without incurring additive dosing effects. Fourth, the shelf also occupies very little hood floor space while framing in a segment of hood floor space where a solid waste container can be co-located with radioactive sample storage space.

The hood layout design also considered post-analysis specimen disposal processing as well.

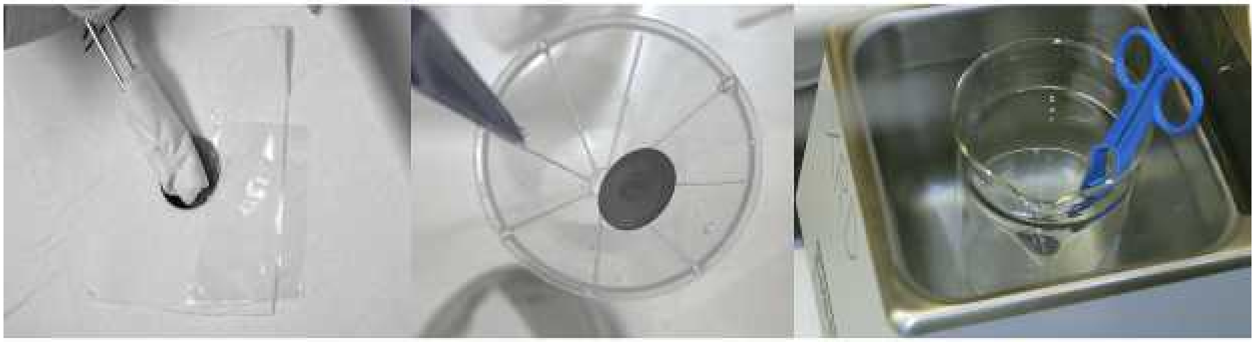
As shown in Figure AIV-1, a hot plate is located in the left rear corner of the hood. During specimen disposal processing of specimen materials, the silicon substrates and aluminum specimen cups are typically cleaned and re-used (combination of parts cost approximately $200 to replace). The assembled specimen is typically disassembled during the cleaning process with the first step involving the separation of the silicon substrate insert from the specimen cup. The parts are placed in a metal slip can lid as indicated in Figure AIV-3. This lid is then placed on the hot plate in order to melt the petroleum jelly adherent used to adhere the substrate to the specimen cup. A piece of tape is attached to the uncoated edge of the substrate using tweezers or other tool per preference of the individual performing the clean-up operations. The tape is used to carefully lift the substrate out of the cup and place it deposit side down on a paper towel. A paper towel-wrapped pair of tweezers is then used for two purposes: (1) to wipe the petroleum jelly adherent off of side facing up, and (2) to push the substrate around on the paper towel to wipe off the deposit materials. The edge of the paper towel coupon can be lifted using tweezers to flip over the silicon substrate. The deposit side must be wiped clean to the standard of "no visible material." The substrate is then transferred into the plastic funnel indicated. This is a re-purposed conventional funnel that makes retrieval of small parts very easy. A plastic clamp is attached to the funnel to aid in the transfer of the funnel into a 600-mL glass beaker located inside the ultrasonic bath shown on the right front side of Figure AIV-1 and the lower right image of Figure AIV-3. The plastic funnel has an additional benefit in that contact between the funnel and the single crystal silicon substrate being cleaned is unlikely to result in damaging chips or scratches on the substrate. This part can then be ultrasonically cleaned while other clean-up activities are performed. After the sonic cleaning process is completed, the funnel can be lifted out of the beaker, carefully drained of all water, and the cleaned parts can be transferred onto a clean paper towel for drying. The sample cup is frequently cleaned by wiping all exterior surfaces with a damp paper towel with subsequent drying. The cleaned pair of parts are then placed into a clean petri dish and labeled as indicated in Figure AIV-4. The petri dish then can be safely stored for future use. Other tools, such as mortar, pestle, tweezers, microspatulas, and acrylic plates, undergo similar cleaning operations prior to being stored for future use with other clean tools.



Place specimen on paper in metal lid, place metal lid on hot plate to melt petroleum jelly adhering silicon substrate to aluminum cup

Apply tape to uncoated edge, lift substrate out of cup

Place deposit side down on paper towel



Paper towel wrapped tweezers wipe the petroleum jelly off the exposed surface and move the substrate relative to the paper towel to remove the powder deposit

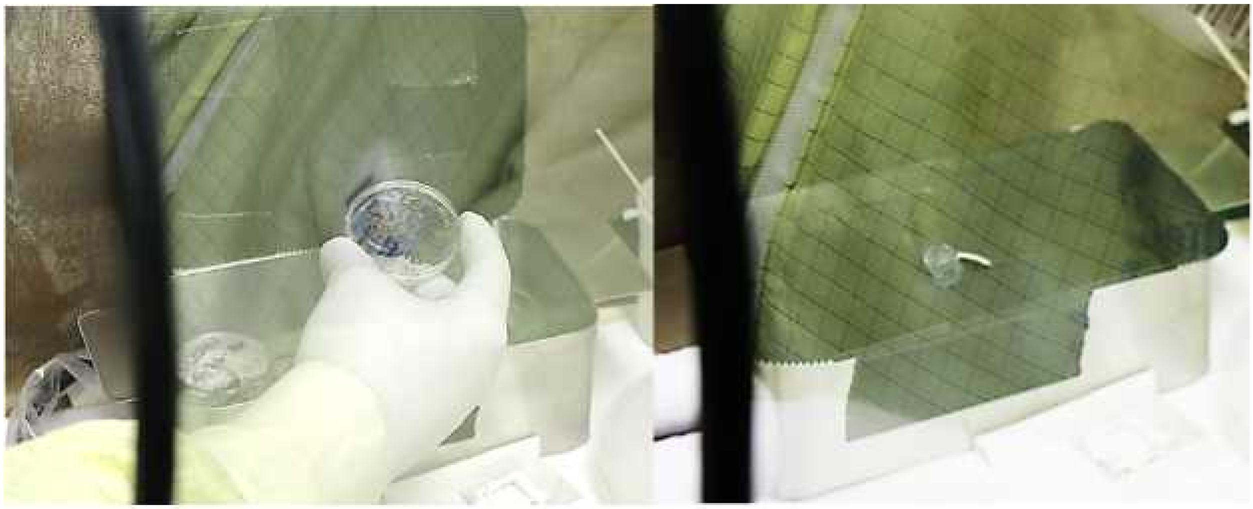
With "no visible" debris left on silicon substrate, place in plastic funnel

Place plastic funnel inside 600mL beaker in ultrasonic water bath (the beaker reduces the volume of waste water generated). Sonicate until clean (few minutes)

**Figure AIV-3. This composite image illustrates the sequence of operations used to safely recover re-useable specimen mount components.**

After wiping the aluminum sample cup thoroughly with a damp paper towel (or sonicating as needed), both parts are dried off and placed in a clean petri dish

Place both clean parts in a labeled petri dish



Place the petri dish in the storage container

Close the storage container to protect the clean components from airborne contaminants

**Figure AIV-4. This composite image indicates the typical sequence of steps used to safely store cleaned specimen preparation components for future use.**