

5.0 RECOMMENDATIONS

Many practical lessons were learned during ICET Test #1 that may serve to improve the quality of information obtained in subsequent tests and the efficiency with which daily operations can be managed. The following items have been discussed with the NRC/industry sponsors, with input from LANL and UNM investigators for consideration as minor modifications to the ICET Test #2 plan and procedures.

- Continue the practice of daily water sampling, but reduce the frequency of comparison between filtered and unfiltered samples if the differences again become negligible. In Test #1, there was no measurable difference in TSS, viscosity, or ICP elemental composition after the bulk turbidity dropped.
- Given the continued increase in the rate of observed precipitation in extracted samples and the continued increase in aluminum concentrations beyond 15 days, plan all subsequent tests for a duration of 30 days.
- Acquire duplicate water baths to improve control of water temperature for extracted samples waiting for viscosity and turbidity measurement. The post-Test #1 interest in controlled-temperature precipitation studies further justifies the need for this equipment.
- Continue the practice of daily water sample viscosity measurements, but eliminate the requirement for replicate measurements if the same level of precision is achieved. Variations between repeated measurements under the Test #1 protocol were less than 1%.
- The presence of deposits on exposed surfaces of the fiberglass blankets and the decline of silicon concentrations in solution raise questions about realistic exposure of fiberglass debris to the test solution. SS mesh sample bags were prepared for Test #1 to confine the fibers, but deposits were noted only on fiber layers next to the mesh, even for mesh envelopes embedded in larger blankets.
 - For Test #2, construct a small mesh sample box (in addition to the original bags) to hold a loose collection of fiber that is not compressed on all sides.
 - Wrap a 1/2-in.- to 1-in.-thick mesh bag around the lower 4 in. of the drain screen to expose a small amount of fiber to higher water velocities.

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6.0 REFERENCES

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6. Ibid 5.
7. Goldstein, J., Scanning Electron Microscopy and X-ray Microanalysis, 2nd ed., Plenum Press: New York (1992).
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18. Murthy, N. Chapter 7, "Materials Characterization and Chemical Analysis," 2nd ed., John Sibilis, Ed., VCH Publishers, Inc., New York (1996).
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21. Ibid 18.
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- 23 U.S. Environmental Protection Agency, "EPA Method 310.1, Alkalinity - Titrimetric, pH 4.5 Alkalinity," EPA Report No. 600/4-79-020.

Preface to Appendices

This preface provides a brief guide to assist in the access and interpretation of the data appendices that follow. Standardized nomenclature is defined first to clarify the origin of samples that are described in the data sets. Then an outline of the appendices is provided with a description of how they were compiled. A list of figures with captions and page numbers is provided for each individual appendix to facilitate access of the desired information.

Nomenclature

Many spatially unique but physically similar sample types were collected in ICET Test 1. To ensure that consistent interpretations and comparisons of data sets are made, it is imperative that a standardized nomenclature be adopted when referring to each sample type. Many different qualitative descriptions of these samples might be equally suitable, but different adjectives convey different connotations to each observer. Therefore, the following definitions establish the convention used in this report when making generic references to sample type. Every effort should be made to adhere to this standard when interpreting the data so that all future audiences will have a common understanding of sample origins from the ICET series.

White Precipitate Upon cooling below the test temperature, T1 daily water samples extracted from the tank formed a visible white material that is referred to as a precipitate. Although the exact formation mechanism has not been confirmed, the material exists as a distinct physical phase separate from the aqueous solution from which it evolved.

Latent Debris Commercial power plants gradually accumulate dust, dirt, and fibrous lint that are generically referred to as latent debris. This classification distinguishes resident material from debris generated during the accident scenario. At the beginning of T1, measured quantities of crushed concrete and soil were added to simulate the latent debris present in containment. These materials were examined via SEM/EDS to establish a baseline composition for comparison to sediment samples (see “Sediment” below).

Sediment Surrogate latent debris particulates and fugitive fiberglass fragments that were initially suspended in water at the beginning of T1 gradually settled to the bottom of the tank to form a layer of sediment. During the course of the test, additional material may have been deposited in this layer. At the conclusion of the test, the sediment layer was recovered as completely as possible.

Sludge At the conclusion of T1, all water was drained slowly from the tank and stored in a large plastic reservoir. Upon cooling, this

liquid effluent also precipitated white material that collected in large quantities at the bottom of the reservoir. Although the material produced at the end of the test is certainly related to the precipitate observed in the daily water samples, it will be referred to as “sludge” to connote the quantities that were produced and to identify the exact source of the samples that were examined.

- White Residue** At the conclusion of T1, all water was drained slowly from the tank. Exposed metal surfaces that cooled rapidly collected a thin deposit of white residue or scale. Some of this material was scraped from internal piping surfaces and tank walls for comparison with other sample types, such as white precipitate from the daily water samples.
- Fiberglass** One of the principal debris types introduced to T1 was shredded fiberglass insulation. This debris was bundled in 3-in.-thick bags (or blankets) made of SS mesh to prevent ingestion through the pump and to better control the placement of debris in various flow regimes. Fiberglass samples are designated by their placement in high-flow and low-flow areas of the tank. Additional, small, 4-in.-square envelopes of fiberglass were also prepared for extraction during the course of the test. These samples are referred to as “sacrificial” samples. Some amount of fiber, especially short fiber fragments, escaped the mesh bags and was deposited in other locations within the tank. This material is referred to as “fugitive” fiberglass.
- Drain Screen** A 10-in.-tall screen made of coarse SS mesh wrapped into a 2-in.-diameter cylinder was inserted into the outlet drain at the bottom of the tank to protect the pump from ingestion of large debris items. Because the drain screen was exposed to higher velocity-directed water flow, it gradually accumulated a layer of debris around the lower few inches of mesh. This material was examined as a separate debris location to identify any apparent differences with other sample locations, such as fiberglass blankets and tank sediment.
- Gelatinous Material** This term generically refers to any observed sample constituent with amorphous, hydrated, or noncrystalline physical characteristics.
- Colloidal Suspension** Several different diagnostic techniques have shown evidence of microscopic particles suspended in T1 test liquid that are invisible to the unaided eye. The extremely small sizes of these particles (5 to 20 nm) suggest that they may be colloidal in nature and that

they remain in suspension or are formed in suspension after larger particulates have settled to the bottom.

- Water Sample** Daily water samples are extracted from the ICET tank for elemental concentration analyses. After properly flushing the sample line, some of this water is extracted directly from the tap. An equal amount of water is also generally collected through a micropore filter. Thus, daily water samples are designated as filtered (F) and unfiltered (U), and a corresponding filter paper exists in the sample archive for each daily sample that is collected.
- High-Volume Filter** In addition to the relatively small volumes collected during daily water sampling, larger quantities were periodically extracted for filtration to determine whether suspended chemical products were present in the test liquid under in situ conditions. The intent of this exercise was to maintain the liquid temperature while forcing the liquid through a micropore filter under vacuum. Temperature control for the T1 high-volume filter samples was not ideal, so the collected filtrate may show evidence of temperature-dependent precipitation similar to that described for white precipitate.
- Filter Paper** Many different samples of tank solution were fractionated by micropore filtration into a liquid supernate and a solid filtrate that existed at the time and temperature condition of the filtering process. These samples include (1) daily water samples filtered during extraction, (2) daily water samples filtered after cooling to room temperature, and (3) high-volume water samples.
- Chemical Deposits** Sacrificial fiberglass samples that were extracted at Day 15 and Day 30 showed evidence of chemical products forming on and between fiber strands. These products are referred to as “deposits,” although the exact physical mechanism of formation is not well understood. The physical appearance suggests growth, agglomeration, or crystallization on and around the fiber strands over time rather than capture or impaction of particles from the bulk solution. This observation is supported by the fact that the small fiberglass samples were located in a region of very low directed water flow (i.e., in the interior of larger blankets).
- Concrete Sample** Several chips of concrete (1/4 in. to 3/4 in. in diameter) were broken from the primary slab of submerged concrete and introduced to the tank in a small SS envelope at the start of the test. Examinations of these chips were conducted to determine if concrete surfaces provide a preferential site for gel formation.

Having defined these terms, the reader may note, nonetheless, minor inconsistencies in the caption labels of these appendices. The caption labels use the same descriptions that were applied in laboratory notebooks to improve traceability of the data.

Usage

Eight appendices are provided that present data collected for the following sample types and analysis methods:

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| (A) | SEM data for 24-hour high-volume filtrate; | p. A-1 |
| (B) | SEM/EDS of the Test-1 Day-15 fiberglass and filtrate; | p. B-1 |
| (C) | SEM/EDS data for Test-1 Day-30 fiberglass and filtrate; | p. C-1 |
| (D) | SEM/EDS data for white precipitate, Day-30 fiberglass, drain screen debris, pipe residue, tank sediment, concrete samples, latent debris baselines, and Test-1 Day-30 high-volume filtrate; | p. D-1 |
| (E) | TEM analyses of Test-1 Day-15 water samples; | p. E-1 |
| (F) | TEM analyses of Test-1 Day-30 water samples; | p. F-1 |
| (G) | TEM analyses of pre-test 1 laboratory solution; | p. G-1 |
| (H) | Sediment Analysis for Test #1. | p. H-1 |

These data are largely qualitative in nature, consisting primarily of SEM and TEM micrographs and EDS spectra. Each appendix represents a separate session of laboratory work that can be traced to a batch of samples that were processed in chronological order. This organization scheme preserves the connection with laboratory notebooks and timelines that naturally developed during operation; however, in a few cases, results for a given sample type may be mixed across two or more appendices because of the order in which the individual samples were analyzed.

Transcriptions of the logbooks are provided for each appendix to document better commonalities that existed among the samples at the time of analysis. Interpretation and understanding of the images and their accompanying EDS spectra will be greatly improved by referring frequently to the logbook sample descriptions and sequences. Typically, a relative large quantity of a test sample was delivered for SEM or TEM analysis, and then several small subsamples of each item were examined. Note that each subsample was assigned a sequential reference number during the laboratory session. These reference numbers have been cited in the figure captions wherever possible to preserve the connection between the micrographs and the notebook descriptions. Electronic filenames have also been stamped on the images to permit retrieval of the original data files that are archived elsewhere. Individual data sets for a given sample item have been collated into a typical sequence of (1) visual image, (2) EDS spectra, and (3) semiquantitative mass composition.

For most of the EDS spectra, semiquantitative mass compositions are also presented. These results are obtained from a commercial algorithm that decomposes the spectra into the separate contributions of each element. Several caveats should be considered when interpreting the numeric compositions thus obtained; however, despite these caveats, semiquantitative EDS analysis offers a natural complement to micrographic examination as a survey technique for identifying trends in composition.

1. The spectral deconvolution algorithm is based on a library of unique signatures of each element that were obtained for pure samples using a standard beam setting that may not match identically the conditions applied for the test item.
2. The operator must select a limited number of elements to be used in the proportional mass balance. These candidates are chosen from among the peaks that are observed in the spectrum; however, the composition percentages can vary, depending on which elements are included in the list. In a few cases, two or more alternative compositions have been generated by selecting a different set of elements from the same spectrum to illustrate the sensitivity of this technique to operator input.
3. The spectral unfolding algorithm is a statistical technique having a precision that depends on the relative quality of the data in each peak. Compositions with high R^2 correlation coefficients and total-mass normalization factors closer to unity represent the more reliable estimates. The precision obtained in the fit depends on the duration of the scan and the number of counts received in each energy bin.
4. All subsamples examined in the SEM microprobe facility are coated with a thin layer of either carbon or gold/palladium alloy to prevent charge accumulation from the impinging electron beam. Spectral peaks visible for gold (Au) and palladium (Pd) are not indigenous to the samples.
5. The EDS spectral analysis software contains a peak-recognition algorithm and an automated cursor that snaps across the spectrum to locate each peak. An accompanying library of elemental energy signatures is also provided to suggest what constituents might be contributing to a given energy bin, but the operator must judge what label to assign to the spectral image. It is possible that some peaks near closely neighboring elements have been mislabeled in these appendices. However, every effort was made to choose from candidate elements that were most likely to be present in the test material. In a few cases, the spectral peaks were not labeled by the SEM operator. These spectra should be viewed as corroborating evidence for similar samples that are labeled in a definitive manner. Careful comparisons of the energy scales in combination with a library of electron scattering energies can also be used to infer the origin of the more prominent peaks that are present in unlabeled spectra.

6. In general, the scan area of an EDS is comparable to $10 \times 10 \mu\text{m}$ unless noted otherwise for a special purpose, such as examination of a 7- μm -diameter fiber strand or obtaining a bulk-average composition over a heterogeneous sample.
7. Unless an obvious spatial heterogeneity is being examined, the exact location of an EDS spectrum is not always relevant because the operator chooses arbitrary sites that are visually judged to be representative. It is not possible to sample a surface comprehensively on a microscopic basis and compute average compositions. In many cases, two or three replicate spectra are provided for this purpose, but SEM/EDS is most effective as a survey diagnostic.
8. For several reasons, EDS analysis is not particularly sensitive to the presence of boron: (a) boron has a low atomic mass that does not interact well with electrons in the beam, (b) the emission lines are very close to those of carbon, and (c) the beam port material has a high absorption cross section for these emission energies. Therefore, the correction factors used in the semiquantitative composition analysis are quite large, as are the uncertainties in the estimated percentage of total composition for this element. There may spectra presented in these appendices where the lowest energy peak is labeled as either B or C when in fact either both are present or the opposite element is present.

EDS locations were chosen manually at regions of specific interest. In many cases, multiple spectra were collected from a single sample and an annotated image is provided to identify the specific location. These annotated images are not generally noted in the laboratory log book entries, but they are provided in proper sequence within the appendices.

Appendices E and F present transmission electron microscopy data for water samples extracted from the ICET solution at Day 15 and Day 30, respectively. The purpose of this examination was to determine whether the physical structure of any suspended products exhibits crystalline or amorphous characteristics. These data are also qualitative in nature, consisting generally of a set of high-resolution micrographs followed by companion electron diffraction images. The TEM sample holder consists of a carbon grid that is “lacey” or filamentary in nature. This grid is visible as a relatively large-scale structure in the background of most images. Surface tension in a droplet of liquid suspends the particulates of interest across the grid so that the electron beam can illuminate the sample through the holes without interference from a substrate. Crystalline material will exhibit diffraction patterns unique to the molecular arrangement. Amorphous material that is diffuse or disorganized in structure will not exhibit regular diffraction patterns that can be identified.

Water samples submitted for TEM analysis are not temperature controlled because the temperature cannot be maintained during the examination. Therefore, the particulates that are observed in these samples are closely related to, if not identical to, the visible white precipitate that is observed in the daily sample bottles at room temperature.

In a few cases, data file names that were noted by the operator in the laboratory log were not successfully saved in electronic form. These cases are noted in the transcribed log sheets, but the corresponding images are unavailable and therefore cannot be presented in the data sequence.

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