Thermal and Chemical Behavior of Uranium-Metal-Bearing Hanford K East Basin Sludge

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Abstract

Uranium-metal-bearing sludge from the Hanford site K East Basin is to be loaded into containers and moved to interim storage in a dry cell at T Plant on the Hanford site. Thermal and chemical behavior of this sludge in its storage container are of interest to design and safety, because oxidation of its uranium generates power and hydrogen gas, with resulting implications for flammability of the container and cell headspaces, and the potential for local temperature escalation. Key aspects of experimental work and model development necessary to support the technical basis for design and safety analyses are reported here: (1) Experimental data supporting the oxidation rate law, including reactive surface area depletion, (2) Experimental data defining the maximum allowable heat generation rate in wet sludge, the so-called dryout heat flux, and (3) Integral modeling of physical and chemical processes to predict temperature and hydrogen concentration histories during loading, shipping, handling, and storage.

Introduction

Metallic uranium Spent Nuclear Fuel (SNF) is currently stored in two water-filled concrete pools, 105-KE Basin (KE Basin) and 105-KW Basin (KW Basin), at the United States Department of Energy (U. S. DOE) Hanford Site, in southeastern Washington State. The Spent Nuclear Fuel Project (SNF Project) is responsible to DOE for operation of these fuel storage pools and for the 2100 metric tons of SNF materials that they contain. These fuel storage pools also contain hazardous substances that primarily result from the degradation of the SNF. The hazardous substances consist of the SNF, sludge, debris, and water.

Both the KE and the KW Basins contain contaminated [i.e., radioactive, polychlorinated biphenyl (PCB), etc.] sludge (~52 m^3). Sludge on the floor and in the pits of the KE Basin is a mix of fuel corrosion products (including metallic uranium, and fission and activation products), small fuel fragments, iron and aluminum oxide, concrete grit, sand, dirt, operational debris and biological debris [1, 2, 3, 4]. The large quantity of fuel corrosion products in the KE Basin floor and pit sludge is a result of the open tops, and in many cases open-screened bottoms, of the fuel storage canisters. Because the SNF stored in the KW Basin was placed in closed containers before storage, most of the corrosion products were retained within the canisters and the sludge buildup in the KW Basin is of much smaller volume than that in KE Basin. The small quantity of sludge on the floor of the KW Basin is assumed to consist primarily of dust and sediment; the floor sludge is not expected to contain significant amounts of fuel corrosion products because the KW Basin canisters have closed tops and bottoms. Sludge in the KE and KW Basin fuel storage canisters (i.e, canister sludge) consists primarily of fuel corrosion products. For the purposes of

differentiating SNF and debris from sludge, any material that will pass through a screen with 0.64 cm (0.25 in.) openings is defined as sludge.

The Hanford K Basin sludge will be managed as two general waste streams: K East (KE) floor, pit, and canister sludge containing relatively low concentrations of fuel particles; and K West (KW) sludge, some of which will contain relatively high concentrations of fuel particles and graphoil. The KE sludge (~43 m^3), which comprises the majority of the total K Basin sludge inventory, will be loaded into Large-Diameter Containers (LDCs) (approximately 5 ft in diameter and 10 ft high) and stored in process cells at T Plant in the Hanford 200 Area. The KW sludge will also be stored at T Plant; however, the storage container configuration has not been specified.

The presence of metallic uranium fuel particles in the K Basin sludge creates the primary technical challenge to the design of the storage systems. The metallic uranium and uranium oxides within the sludge will corrode, hydrate, and, consequently, generate heat and hydrogen gas during storage. Additionally, heat is also generated within the sludge by radiolytic decay. To maintain thermal stability, the sludge must be retrieved, staged, transported, and stored in systems designed to provide a rate of heat removal that prevents the temperature in the sludge from rising above established limits (currently defined as below the sludge boiling point).

This paper describes experimental testing and modeling performed to determine and predict (1) the uranium metal content of KE sludge, (2) limits to heat removal from sludge, and (3) integral modeling to use the experimental information to deduce process conditions and safety limits. All work described here was performed under contract to Fluor Hanford by Pacific Northwest National Laboratories (PNNL) and Fauske & Associates, Inc. (FAI).

Gas Generation Testing to Determine Uranium Metal Content and Particle Size

Gas generation testing with sludge was conducted by the Pacific Northwest National Laboratory (PNNL) to better understand the quantity and reactivity of the metallic uranium present in the K Basin sludge. There are no direct analytical methods for determining the uranium metal content in sludge that includes high concentrations of various uranium oxides. Therefore, the uranium metal content was determined indirectly by oxidizing the sludge samples and measuring the offgas composition and quantity. Delegard et al. [5] provides a detailed description of this work. Specific objectives of this testing included:

1. Determine the quantity of uranium metal present in each sludge type.
2. Estimate the uranium metal surface area per unit mass of sludge Determine if the oxidation rate measured for uranium in actual sludge agrees with the baseline project-accepted kinetic model for uranium oxidation in water [6].
3. Provide data for input into sludge thermal stability models for safe sludge handling practices (e.g., uranium metal content, reaction rates).
4. Provide gas generation data that can be used as design input into sludge transportation and storage systems.

Gas Generating Testing Experimental Approach

The sludge used for the gas generation testing was taken from the K E Basin floor and fuel canisters in March and April 1999 by Duke Engineering & Services Hanford. A consolidated sampling technique was employed for collecting the material (i.e., sludge from several locations was combined to form "consolidated samples"). Three sludge samples were used: fuel canister sludge (KC-2/3); floor sludge collected from between slotted fuel canisters containing highly damaged fuel (KC-4); and floor sludge collected away from fuel canisters and away from areas known to contain high concentrations of organic ion exchange resin (KC-5). The canister sludge used in this testing (KC-2/3) was prepared by combining two consolidated sludge samples (i.e., KC-2, collected from canisters containing highly damaged fuel, and KC-3, collected from canisters containing moderately damaged fuel). Portions of these samples were sieved to separate particles greater than or "plus" 250 µm (P250) from particles less than or "minus" 250 µm (M250). This separation was made to mimic the separation operations that are planned for the retrieval of certain K Basin sludge types and to gain a better understanding of how uranium metal is distributed in the sludge. [The separation point for certain K Basin sludge types was subsequently changed from 250 µm to 500 µm [4].] Fine uranium metal particles have a high surface area and will react rapidly. Larger uranium metal particles will react at a slower rate, since their surface area per unit mass of sludge is lower.

For the testing described here, sludge samples were placed into four large-scale vessels (850 ml) and eight small-scale reaction vessels (30 ml). Each vessel has a separate pressure transducer on the gas manifold line. The entire surface of the reaction system exposed to the sludge sample is stainless steel, except for a gold-plated copper gasket sealing the flange at the top of the reaction vessel.

Figure 1 illustrates the small-scale reaction vessel and shows where the thermocouples are placed inside and outside the vessel. The reaction vessels are 304 stainless steel cylinders, each internally ~1.75 cm diameter and 14 cm high (internal volume ~33 cm^3). For the gas generation testing, each vessel was wrapped in heating tape and insulated. Two thermocouples were attached to the external body, one for temperature control and one for over-temperature protection. Two thermocouples were inserted through the flange. The thermocouple centered in the lower half of the vessel monitored the temperature of the liquid phase; the one centered in the upper half monitored the gas phase temperature within the reaction vessel. The reaction vessels were placed in a hot cell and connected by a thin (0.0058-cm inside diameter) stainless steel tube to the gas manifold outside the hot cell. A stainless steel filter (2-µm pore size, Nupro) protected the tubing and manifold from contamination. A thermocouple was attached to this filter as well. The arrangement of the large-scale reaction vessels was similar to that of the small-scale vessels.

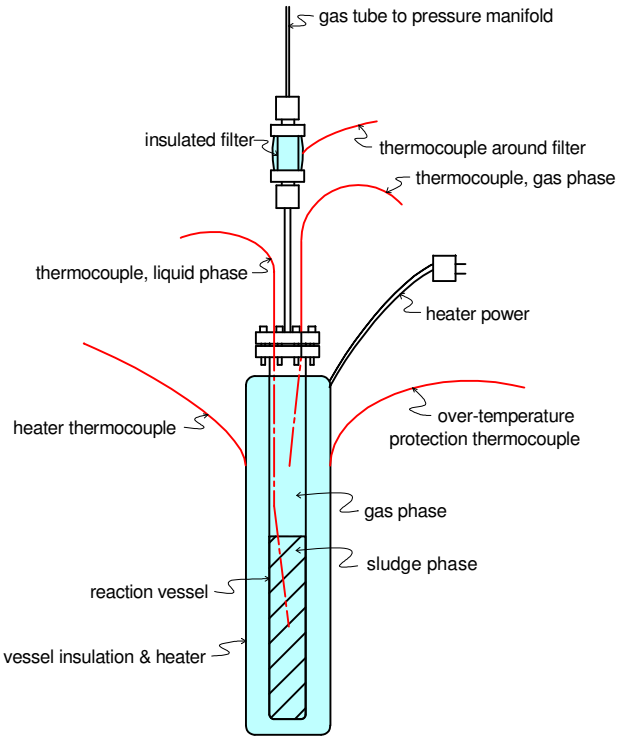

Figure 1. Reaction Vessel Used in Small-Scale Gas Generation Tests.

Temperatures and pressures were recorded every 10 seconds on a datalogger; the data were averaged every 20 minutes and saved in a computer file. Temperature and pressure data were also manually logged once each day. Gas samples were collected intermittently and analyzed via mass spectrometry. An inert cover gas was required to identify product gases and understand the chemical reactions occurring in the settled sludge. Because neon leaks more slowly than helium from the system, it was used as a cover gas. Argon was not used because it served as an indicator of atmospheric contamination. Nitrogen can react with sludge and thus was not a suitable cover gas. The neon was analyzed independently by mass spectrometry and determined to contain no impurities in concentrations significant enough to warrant correction.

At the start of each run, each system was purged by at least eight cycles of pressurizing with neon at 45 psi (310 kPa) and venting to the atmosphere. The systems were at atmospheric pressure, about 745 mm Hg (99.3 kPa), when sealed. The vessels then were heated, and the temperature set points were adjusted to keep the material within 1°C of the desired liquid phase temperatures. For the reactions that were heated, the gas phase temperatures were measured to be 5°C to 25°C lower than those of the sample liquid phase.

Each large-scale vessel contained 70 to 440 grams of settled sludge held at ambient hot cell temperature (~32ºC). The large-scale-test conditions are expected to be prototypical of those that will be experienced during long-term storage of the sludge at T Plant (i.e., large-scale tests serve as a mock-up for prolonged T Plant storage). The small-scale tests, which were conducted with about 15 grams settled sludge each, were conducted at elevated temperatures (six tests at 80ºC, one at 60ºC, and one at 40ºC) to accelerate the reactions and provide conclusive gas generation data within a reasonable testing period. The temperature in all of the small-scale test vessels was

increased to 95°C for a period at the end of the tests to force completion of the reactions. Table 1 below shows the test matrix.

Table 1. Experimental Matrix for Large- and Small-Scale Gas Generation Tests.

Temperature and Test Scale	KC-2/3		KC-4		KC-5	
	M250 µm	P250 µm	M250 µm	P250 µm	M250 µm	P250 µm
40ºC-small		X[(a)]				
60ºC-small		X[(a)]				
80ºC-small	X[(a)]	X[(a)]	X[(a)]	X[(a)]	X[(a)]	X[(a)]
Ambient (~32ºC)-large		X	X X (Dup)		X	

Note: Large-scale tests at ambient temperatures (~32ºC) are being conducted with the >250 µm fraction of KC-2/3 Composite and the intact (before sieving) samples from KC-4, KC-4 Dup, and KC-5.
(a) Tests completed at 95°C.

Gas Generation Testing Results

Small-Scale Gas Generation Testing with Canister Sludge (KC-2/3 P250)

Small-scale gas generation tests were conducted with KC-2/3 P250 at three temperatures: 40ºC, 60ºC, and 80ºC. Each test was then subjected to a period of heating to 95°C to force completion of the reactions. The KC-2/3 P250 sample, which is more reactive than the other samples tested, exhibited a gas generation profile (moles of gas generated as a function of time) that includes an induction period, a linear region, and a declining region as reactants are depleted (Figure 2). The time of the induction period increased with decreasing reaction temperature.

In the linear region at 80ºC, gas was generated at a rate of 1600 ml total gas [at standard temperature and pressure (STP)] per kg settled sludge-day**.** A 27-hour induction period was observed in this test**.** During the first 730 hours for the test conducted at 80ºC, about 95% of the initial uranium metal was depleted, and the composition of the gas ranged from about 75% to 85% H_2 and 15% to 25% CO_2**.** Small quantities of other gases (e.g., methane, ethane, krypton, and xenon) were also present.

Several actions were taken to obtain additional confidence in the data generated from the KC-2/3 P250 test**.** After the gas generation rate slowed down, the KC-2/3 P250 reaction vessel was agitated aggressively for about 20 minutes with a high-energy vibrator. After this agitation, the gas generation quantity and rate were essentially unchanged. Also, near the end of the test, the temperature was raised from 80ºC to 95ºC. The temperature change had little impact on the gas generation quantity or rate. Since these perturbations had little effect on the gas generation, it is unlikely that any metallic uranium particles were somehow shielded (e.g., suffered mass transfer resistances from coating of corrosion products) from participation in the measured corrosion reactions.

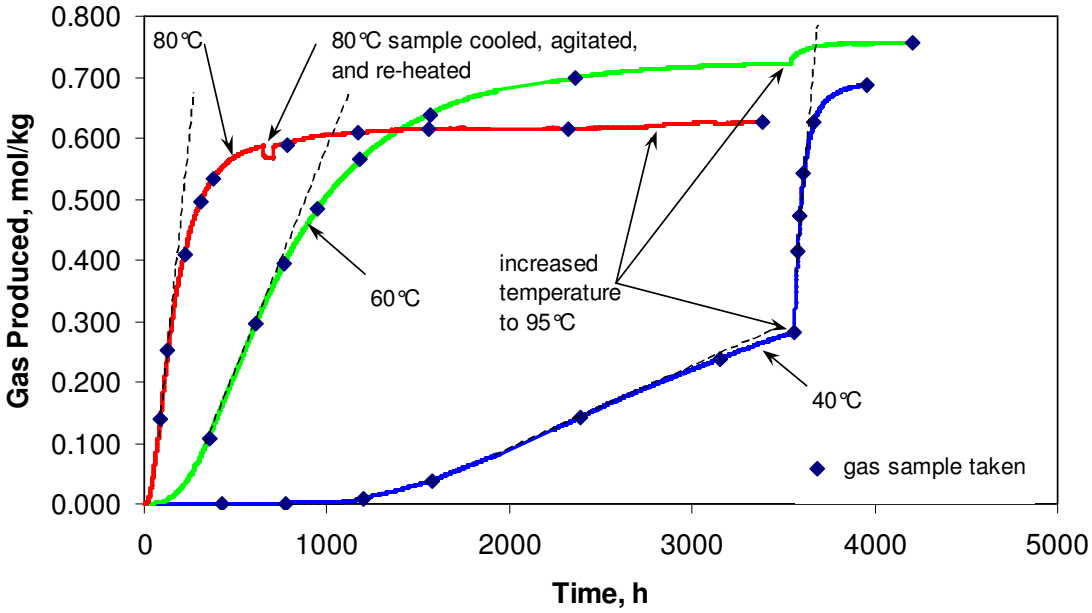

Figure 2. Total Gas Generation from KC-2/3 P250 Material in Small-Scale Reaction Vessels at 40°C, 60°C, and 80°C and Completed at 95°C (dashed lines indicate rates at linear gas generation).

The gas generation rate profile for the test conducted at 60ºC shows that the induction period was about 205 hours (~9 days), and that after 26 days, the reaction was near the end of the linear region. Subsequent heating at 95ºC after 145 days at 60ºC produced only about 5% additional gas. In the linear region at 60ºC, gas was generated at a rate of 440 ml total gas (at STP) per kg of settled sludge-day. In the linear region, the composition of the gas samples was 96% H_2 and 4% CO_2. The H_2 generation rate in the linear region at 60ºC was about a factor of 3 less than the rate obtained in the linear region at 80ºC.

At 40ºC, the reaction induction period was about 1340 hours (~56 days). About 80 ml total gas (at STP) per kg of settled sludge-day were generated in the linear region. Analysis of the gas collected in the linear region showed approximately 1% CO_2 and 99% H_2.

After about 150 days at 40ºC, the reaction vessel temperature was increased to 95ºC. Within several hours, a linear range of gas generation was established at 95ºC. The total gas generation rate at 95ºC was about 4600 ml total gas (at STP) per kg settled sludge-day, and the gas composition was about 98% H_2 and 2% CO_2.

Using the hydrogen gas generation rate data collected during the linear region at 40ºC, 60ºC, 80ºC, and 95ºC, the activation energy was calculated for the reaction:

$$U + 2\ H_2O \rightarrow UO_2 + 2\ H_2$$

The activation energy for uranium metal corrosion was 15.1 kcal/mole (63 kJ/mole). This is near the value for uranium metal corrosion in H_2-saturated water of 16.1 kcal/mole (67 kJ/mole), as derived from a review of the technical literature [7].

The corrosion of uranium metal could be established not only by the observed evolution of hydrogen gas but also by the appearance, in the correct proportions, of krypton and xenon gases of the appropriate isotopic compositions. Furthermore, methane, ethane, and other hydrocarbons in the correct concentrations were observed in the evolved gases from the reaction of uranium carbide traces in the metallic fuel.

Small-Scale Gas Generation Testing with Floor Sludge (KC-4 P250, KC-4 M250, KC-5 P250 and KC-5 M250) and Canister Sludge (KC-2/3 M250) at 80ºC and 95ºC

The rates of gas generation from the tests with KC-4 and KC-5 and KC-2/3 M250 sludge conducted at 80ºC were very low compared with the 80ºC test with KC-2/3 P250 sludge. After 18 days, the gas generation rates were between 4 ml and 25 ml total gas (at STP) per kg of settled sludge-day. Of the gas generated, more than 90% was CO_2.

The finding that KC-2/3 M250 had low reactivity was unanticipated. Its counterpart, KC-2/3 P250, was found to be quite reactive. Furthermore, other data show that the dry particle density of KC-2/3 M250 is much greater than that of KC-2/3 P250 (7.57 g/cm^3 for M250 and 2.23 to 6.91 g/cm^3 for P250), which suggests a higher concentration of dense uranium metal. The total uranium (of all chemical forms) also is higher for the wet sludge M250 fraction, 40 wt% versus 18 wt% for the P250 fraction. The knowledge that uranium metal particles in the canister sludge are generally confined to sludge particles greater than 250 µm will eliminate regions of concern on thermal stability maps generated from modeling efforts.

The reaction vessels were agitated with a high-energy vibrator for 5 minutes each to determine if the sludge reactions with water are inhibited by coatings, films, or other mass transfer resistances surrounding the uranium metal particles. After agitation, the rate of gas generation initially increased and then appeared to return to a rate similar to that exhibited before agitation. Although the magnitudes of the gas generation rate increases are rather small, the gas generation mechanism(s) instigated by the agitation are unknown.

Large-Scale Gas Generation Testing at Ambient Temperature (~32ºC)

Gas samples were collected and analyzed from three large-scale tests: KC-4, KC-4 duplicate, and KC-5. The rates of gas generation from these tests were very low, less than 0.05 ml total gas (at STP) per kg settled sludge-day. The primary gases were CO_2 (63% to 92%) and H_2.

While the total moles of gas generated were nearly identical for KC-4 and KC-4 Dup, the compositions initially varied significantly: 45% H_2 for KC-4 and ~1% H_2 for KC-4 Dup. The compositions became closer with longer reaction time and subsequent gas sampling with neon purging. Although both systems were purged with neon gas multiple times before beginning the run, KC-4 Dup contained about seven times more residual air than KC-4. Oxygen in the system strongly affects the controlling mechanism for uranium metal/water reactions.

The overall gas generation rate of the KC-2/3 P250 large-scale test run at the hot cell temperature was almost 40 times higher (~0.9 ml/kg settled sludge-day) than the rates observed for the KC-4 and KC-5 tests. The gas collected after about 19 days of reaction was about 92% CO_2; about 95% CO_2 was observed over the 195-day test duration. The KC-2/3 P250 large-scale test evidently has not transitioned to the hydrogen-mediated uranium corrosion mechanism even after 195 days of testing at the 32ºC hot cell temperature. The longer induction time at 32ºC likely is because of the larger vapor/sludge volume ratio in this large-scale test (~10) compared with the small-scale test (~1).

Uranium Metal Content in Sludge

From the gas generation data and the projected corrosion reactions, the quantities of metallic uranium initially present in the sludge from each test were calculated. The KC-2/3 P250 sludge contained 7.4±0.9 wt% metallic uranium (on a settled-sludge basis). [This projected uranium metal content is consistent with results obtained from dissolution enthalpy (calorimetry) testing performed on KC-2/3 P250 [8].] All other sludges evaluated in the gas generation testing were estimated to contain less than 0.2 wt% metallic uranium (settled-sludge basis).

Canister sludge sample KC-2/3 was split into two fractions using a 250-µm sieve. Approximately 25% of the original sample was made up of particles greater than 250 µm. Therefore, the projected uranium metal content in the original KC-2/3 canister sludge sample was: 1.9±0.2 wt% [(0.25x7.4±0.9) + (0.75x0.0485)]. [Note sample KC-2/3 M250 was estimated to contain 0.0485 wt% metallic uranium.]

Fission Product Gases

The existence of the krypton and xenon fission gases in the product gas gives evidence of the corrosion of uranium metal, because until corrosion occurs, the fission gases remain trapped within the uranium metal matrix. The krypton and xenon isotope ratios observed in the product gas are similar to the ratios that are expected based on burn-up calculations by the ORIGEN code, which provides a general confirmation to the uranium metal content in the sludges calculated from the gas generation data.

Evaluation of Reaction Surface Area of Uranium Particles in KC-2/3 P250

The rate at which uranium metal reacts with water depends on the available surface area of the uranium particles. An initial evaluation of the uranium metal reaction surface area per unit mass of settled sludge was performed using the results from the gas generation testing with sample KC-2/3 P250 (at 80ºC). Results from the preliminary evaluation indicate that the uranium metal surface area calculated from the gas generation data agrees reasonably well with theoretical predictions using the rate equation given in the Spent Nuclear Fuel Project Technical Databook [6]. [Note: The rate equation in Reilly [6] is being used within the thermal stability models for sludge transportation and storage.]

The uranium metal corrosion in the KC-2/3 P250 sludge sample appears to be well modeled as spheres or cubes of about 800-μm axial dimension based on the rate equation in Reilly 1998. Figure 3 shows an example comparison of test data to a shrinking spherical particle model. This example is typical of observed behavior and is phyically expected. Use of a reaction rate enhancement factor with the rate equation in Reilly [6] is probably not necessary.

Conclusions from Gas Generation Testing

The conclusions resulting from the gas generation tests are summarized as follows:

- The floor sludge has low chemical reactivity, provides little gas generation, and is estimated to contain less than 0.15 wt% metallic uranium (settled-sludge basis).
- Sludge made up of fine particles (<250 μm) from both the KE floor and canisters also has low chemical reactivity and gas generation.
- The canister sludge (including <250 μm and > 250 μm fractions) is estimated to contain 1.9±0.2 wt% metallic uranium (settled-sludge basis).
- Larger-particle canister sludge (>250 μm) exhibits moderate chemical reactivity and gas generation (i.e., contains some metallic uranium).
- The reaction rates observed for larger particle canister sludge agree reasonably well with the rate equation for uranium metal with oxygen-free water given in the Spent Nuclear Fuel Project Technical Databook [6]. Use of a reaction rate enhancement factor is probably not necessary.

Dryout Heat Flux of Liquid-Saturated Sludge

The presence of reactive metal particles in the sludge in a large diameter storage container (LDC) raises the issue of a chemical runaway reaction. From a thermodynamic point of view a runaway reaction can not result in sludge temperatures above 96°C. At 96°C the heat generated by the metal water reaction is equal to the latent heat carried away by water evaporation. A key question in this regard is whether or not interstitial water can be supplied to the runaway reaction zone at a rate sufficient to keep up with the water evaporation rate in the reaction zone, that is at a rate sufficient to prevent dryout in the reaction zone. An experimental study was undertaken to determine the dryout heat flux in a water-saturated sludge [9]. If the chemical reaction power per unit area exceeds the dryout heat flux, the reaction zone would dry out because of an insufficient supply of water.

First-of-a-kind data were obtained for dryout of evaporating water-saturated sludges. The apparatus utilized for the measurement of dryout heat flux data is shown photographically in Figure 4. In general, the apparatus consists of a heat source, a spiral glass condenser and coolant circulation system, and instrumentation for thermocouple readout.

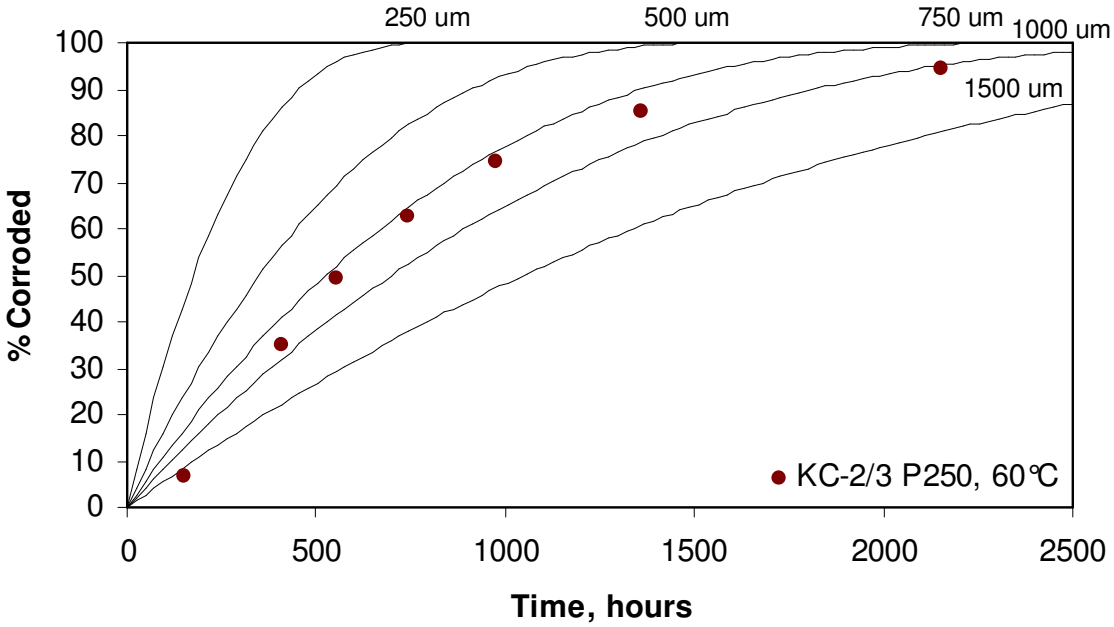

Figure 3. Uranium Metal Corrosion in Water Observed in the KC-2/3 P250 Test at 60°C Compared with Uranium Metal Sphere Corrosion Rates Under Similar Conditions.

A 1000 watt hot plate provides heat to the bottom of the Kaolin simulant sludge. The test section is a Pyrex™* glass pipe having an inside diameter of 10.49 cm and a length 40.6 cm. It is secured at the bottom to a 1.27-cm thick copper plate and at the top to a flange constructed of garolite (cotton fabric/phenolic resin compound) fitted with an adapter to a standard quartz taper fitting. The bottom of the condenser is inserted into the fitting and thereby open to the upward flow of steam from the test section. The condenser coil has an inside diameter of 6.0 mm and an overall length of 120 cm. Cooling water is supplied to the glass condenser directly from the house line. A valve and a flow meter are used to control and measure the cooling water flow rate. Thermocouples are used to monitor the inlet and outlet coolant temperatures at the condenser, the temperature of the copper plate which supports the column of simulant sludge, and the temperatures in the simulant sludge at a height of 1.27 cm above the copper plate and about 1.0 cm below the surface of the simulant sludge. The thermocouple in the copper plate is located in the center of the plate about 0.64 cm below the simulant sludge.

Kaolin simulant is placed in the bottom of the test section and water is added to create a layer about 10% of the sludge height, similar to the overlying water expected in a sludge storage container. The outside of the test section and condenser are wrapped in insulation; observations were made to ensure that all steam generated in the test section is captured in the condenser coils. It takes about 5 hours to uniformly heat the sludge column to the point of incipient dryout. The dryout limit is measured by noting the temperature difference between the inlet and outlet of the condenser. If, following an increase in heat added to the sludge column, the temperature difference remains approximately constant for several minutes, the heat flux is significantly increased to determine if a true dryout has been reached. If, after another several minutes, the

* ™: Laboratory Equipment Corp.

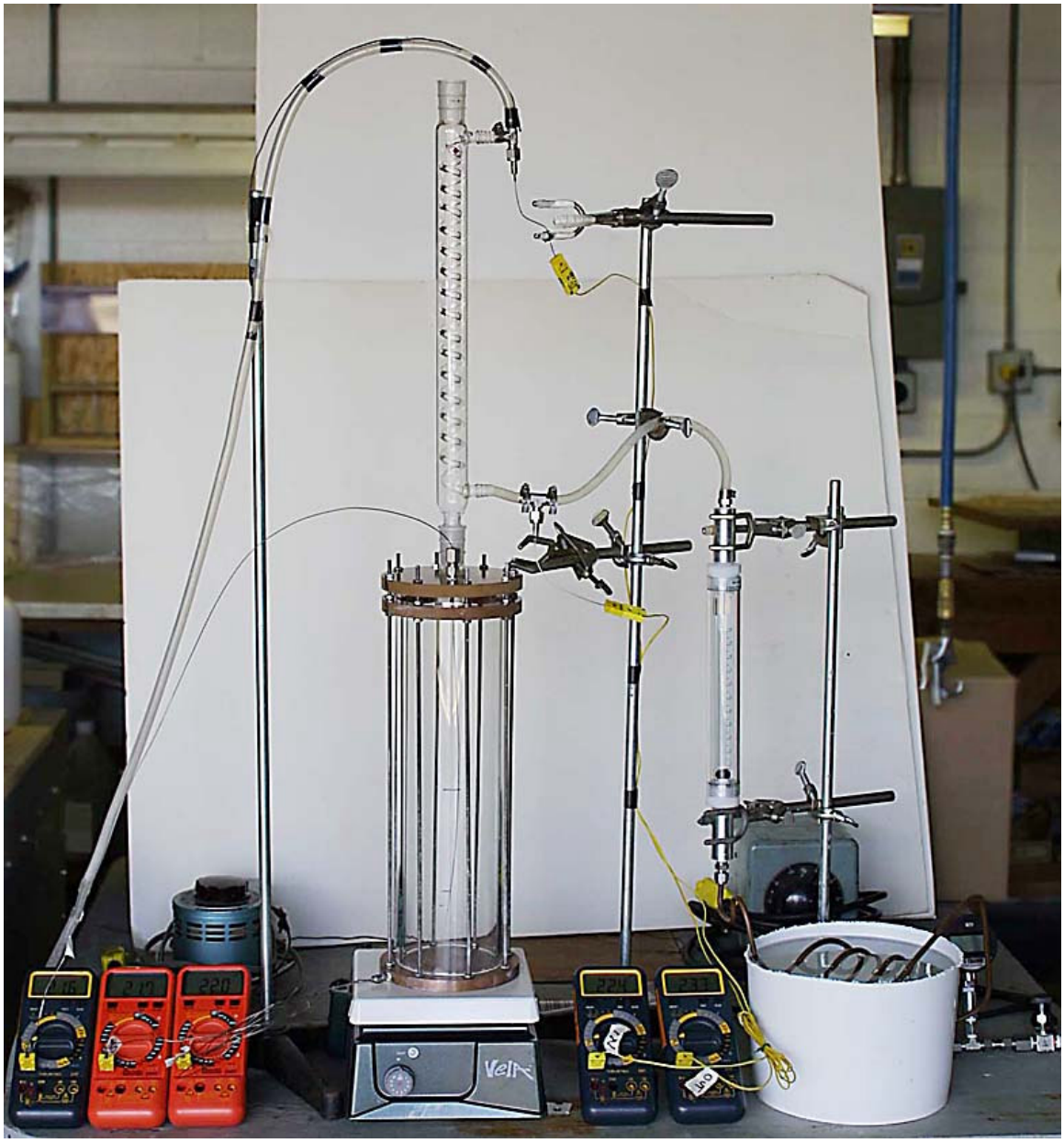

Figure 4. Sludge Dryout Heat Flux Experimental Apparatus.

temperature difference measured across the condenser remains essentially the same or decreases, the maximum temperature difference observed during this time is taken as corresponding to incipient dryout. The dryout heat flux is calculated from the heat balance applied to the condenser-coolant flow using this temperature difference.

Dryout heat flux data were taken for two different mixtures of water and kaolin: 55 wt% kaolin (350 Pa yield strength) and 60 wt% kaolin (10^3 Pa yield strength). Three tests were performed with the 55 wt% kaolin mixture and five tests were performed with the 60 wt% kaolin mix. The sludge depths were approximately 5.0, 10.0 and 16.0 cm.

Conditions and results for the eight experiments performed are summarized in Table 2. Data indicate that the dryout heat flux is independent of sludge depth and sludge strength. This behavior can be successfully predicted using a porous medium model with the gravitation head difference between water and steam providing the driving force for water flow, balanced by frictional loss in the porous medium:

$$\dot{q}''_{dry} = \frac{(\rho_f - \rho_g) g\, h_{fg} \kappa}{\nu_f}$$

where $\dot{q}''_{dry}$ is the dryout heat flux, W/ m^2, ρ_f = 1000 kg/m^3 and ρ_g (negligible) are water and gas densities, g = 9.8 m/s^2 is the acceleration of gravity, h_{fg} = 2.2 x 10^6 J/kg is the heat of vaporization, ν_f = 2.98 x 10^{-7} m^2/s is the liquid viscosity, and $\kappa = 10^{-13}$ m^2 is the permeability for strong clay materials. Inserting these values yields $\dot{q}''_{dry}$ = 7235 W/m^2. As can be seen from Figure 5, this prediction is close to the measured dryout heat flux values.

The dryout heat flux model can be applied to a hypothetical situation in which substantial segregation of the reactive uranium metal has occurred in a sludge container. Using the safety basis metal content derived from the gas generation experiments described above, even if all the metal segregates into a single layer, the dryout heat flux is not exceeded. This hypothetical situation is unrealistically pessimistic based upon models [10] and simulant experiments [11] for settling of sludge. Therefore, using the combination of gas generation and dryout heat flux experimental data, it is a priori known that K East sludge will be unconditionally thermally stable in its planned storage container and environment.

Conclusions from Sludge Dryout Experiments

Conclusions of the sludge dryout experiments are:

(1) The measured dryout heat fluxes fall within the narrow range 7400 to 10,300 W m^{-2}, regardless of the depth of the sludge column or the strength of the sludge.

(2) The dryout heat flux is compatible with a model based on gravity-driven counter flow of water and steam within the sludge column. In the model steam rises through particle-free cracks that penetrate the sludge column. The liquid flows downward through regions of particle-rich saturated sludge. The liquid flow controls the dryout heat flux.

(3) Interestingly enough, while consolidation does not appear to aid the downward flow of liquid to the hot boundary at the bottom of the sludge column, it seems to prevent the sludge just above the bottom from drying out after the dryout heat flux is exceeded. This behavior is quite different from that in nonconsolidating packed beds from which the water is expelled during the post dryout period.

Table 2. Sludge Simulant Dryout Heat Flux Data.

Test	Wt.% Kaolin	Simulant Sludge Yield Strength (Pa)	Sludge Depth (cm)	Coolant Flow Rate (L min^{-1})	Temperatures at Dryout (°C)				Dryout Heat Flux (W m^{-2})
					Sludge Bottom	Sludge Top	Condenser Inlet	Condenser Outlet	
1	55	300	5.3	0.164	99.7	99.2	22.6	28.6	7930
2	55	300	10.5	0.161	99.1	98.7	23.8	30.9	9220
3	55	300	16.0	0.160	99.2	98.9	22.8	30.8	10300
4	60	1000	6.0	0.160	99.5	99.0	25.0	32.5	9680
5	60	1000	5.0	0.164	99.2	98.5	23.4	29.0	7430
6	60	1000	5.1	0.164	99.4	98.9	25.1	32.8	10300
7	60	1000	10.6	0.156	99.7	99.0	23.3	29.2	7430
8	60	1000	15.5	0.170	99.4	98.7	23.0	28.7	7820

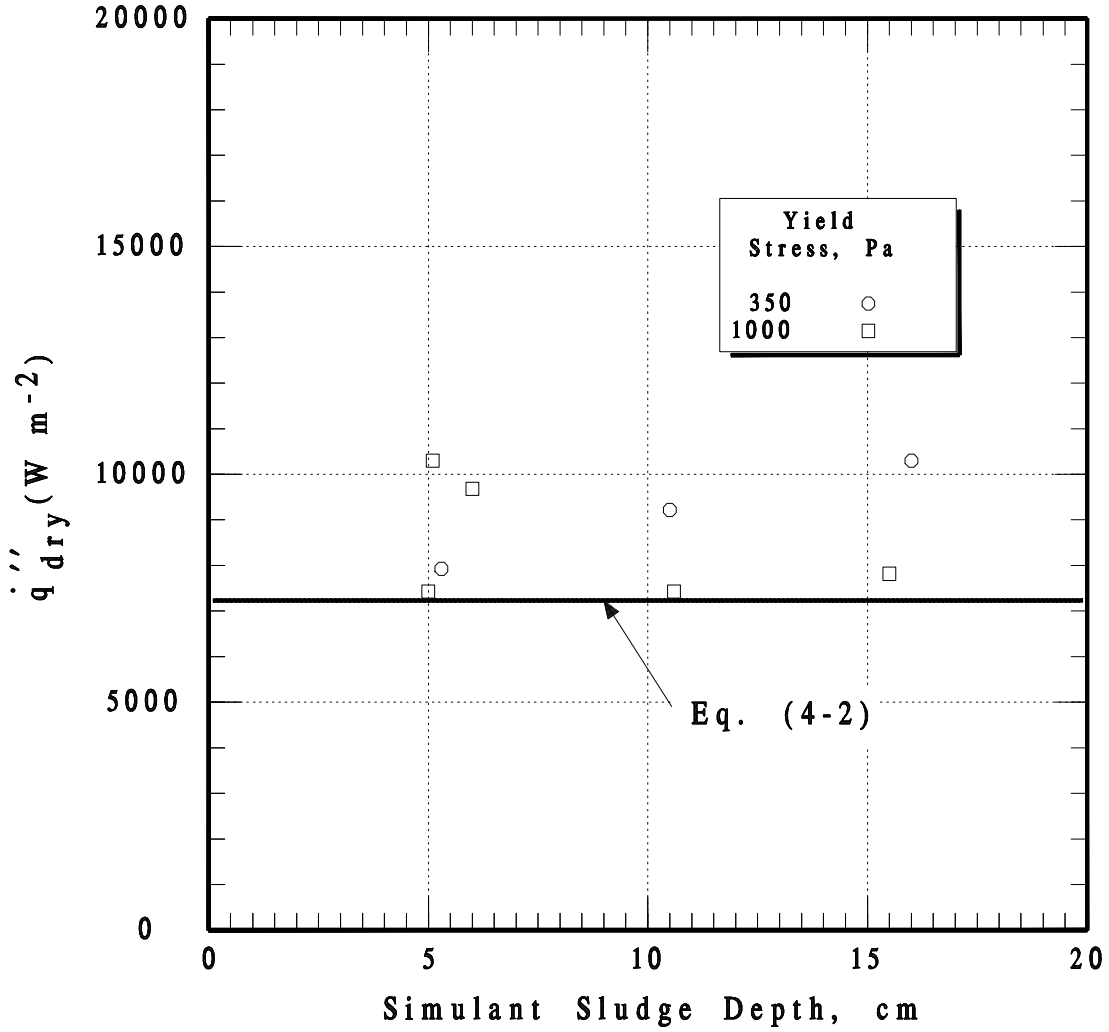

Figure 5. Sludge Simulant Dryout Heat Flux Data Compared to Prediction (Indicated Equation Number is from Reference).

(4) All physical uranium metal concentrations and arrangements within sludge columns produce reaction heat fluxes that are less than the dryout heat flux. Thus a runaway metal/water reaction in a sludge column will not result in local sludge temperatures above T_{eq} = 96°C. This is true for K East sludge but this work has not yet been applied to K West and fuel piece sludge.

Integral Modeling of K East Sludge Thermal and Chemical Behavior

While experiments described above prove conclusively that K East sludge will be thermally stable in their planned storage container and environment, integral modeling of sludge thermal and chemical behavior is necessary to understand safety margin and to analyze off-normal scenarios. A thermal analysis of record [12] used a detailed finite-element conduction model of the

storage container, the LDC, to predict that sludge temperatures remained stable during loading, storage, and transportation. However, this conduction analysis could not consider physical limitations to sludge heat flux as found in the experiment. Also, an iterative approach was required between the finite element model and a simple building model to establish consistent conditions for storage in a T Plant cell.

A general model created for Hanford spent nuclear fuel and waste facility analyses, HANSF, was used to scope design options for the LDC and provide an independent, a priori evaluation of the sludge maximum temperature [13]. The benefits of this model are flexibility for design calculations, coupling of fluid flow and heat conduction to simplify analyses of an LDC in a storage facility, capability to perform accident analyses, and aerosol transport and deposition models to predict facility leak path factors.

Briefly, HANSF can model heat transfer, fluid flow, and chemical reactions in sludge, its container, a cask, if present, a building or facility containing them, and the environment. Decay power, oxidation power, conversion of metal to oxide, and reduction of reactive surface area are included. Heat conduction in sludge and its container is allowed in one or more dimensions, according to the problem; natural convection occurs in overlying water or air. Pressure, temperature, gas composition, and exchange flows are considered in control volumes that typically consist of the container headspace and surrounding compartments. Aerosol phenomena and combustion can are treated for off-normal calculations such as container overpressurization scenarios. HANSF was developed and is maintained under the Fauske & Associates, Inc. NQA-1 compliant Quality Assurance program.

Capabilities described here are easily generalized to other sludge and fuel containers, and are routinely applied to other facilities such as vacuum drying, shipping, and storage of Hanford Spent Nuclear Fuel, Hanford tank farms, and the Hanford Waste Treatment Plant.

A typical model of the LDC is shown in Figure 6. Two-dimensional conduction occurs in sludge and its container, and convection occurs within the overlying liquid and headspace gas. Open ports to the surroundings allow prediction of natural circulation rates. One or more containers may be modeled in a storage cell (Figure 7) so that heat transfer and fluid flows in the LDC and facility are fully coupled.

Results for a limiting case of LDC storage in a T Plant cell with loss of forced ventilation are shown in Figure 8. Hydrogen concentration (Figure 8, upper left) in the cell reaches a peak value of 2%, achieved by laminar exchange flow through gaps in the cover blocks separating the cell from the overlying canyon (Figure 8, lower right). Sludge temperature reaches a peak value of about 66 C, and its decline is due to a loss of reactive surface area as uranium metal particles shrink.

This model was used to size ventilation ports from the LDC so that at the time of peak hydrogen generation within the LDC, the hydrogen concentration was below the lean flammability limit in both the container and cell.

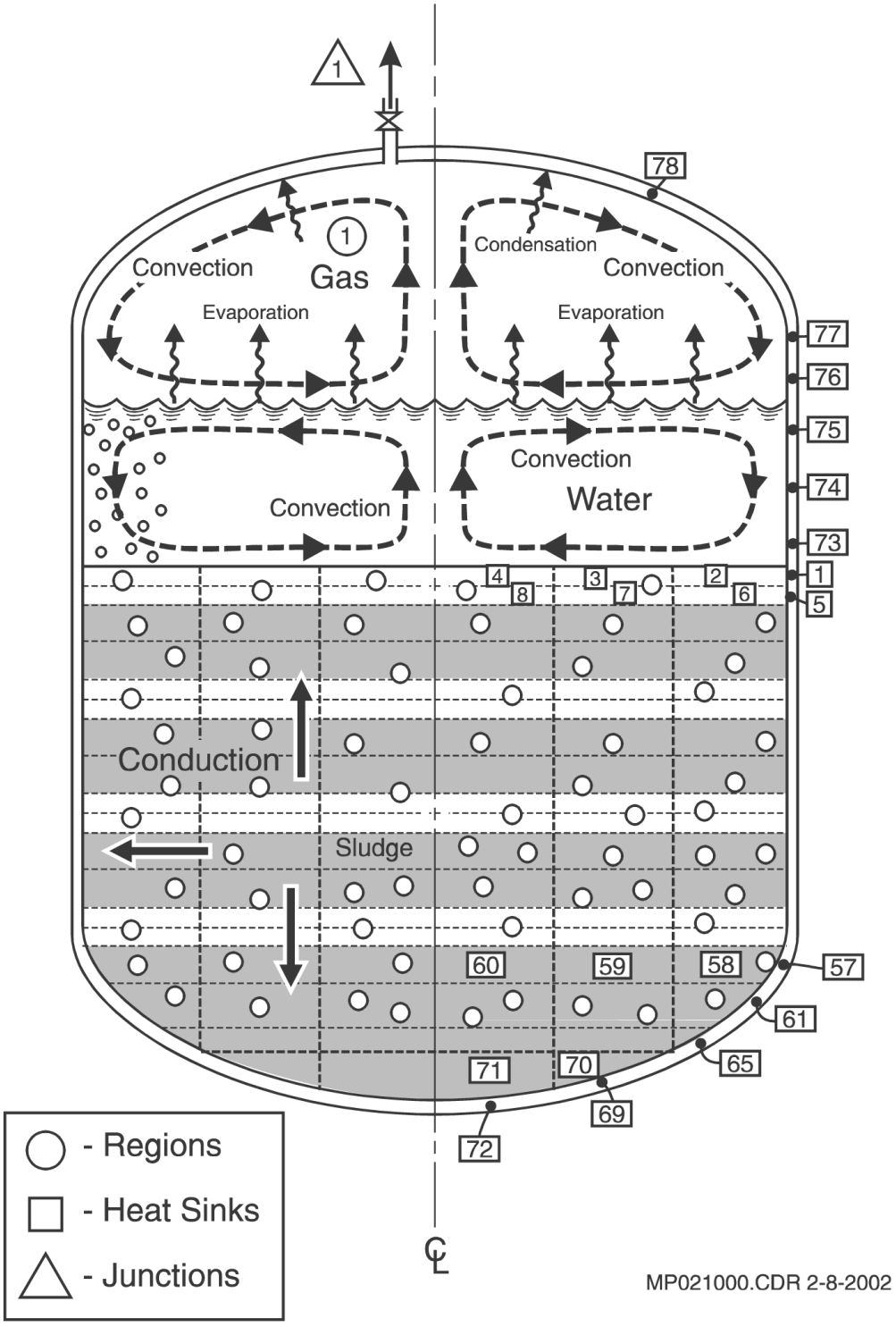

Figure 6. Nodalization and Physical Processes for Sludge and LDC.

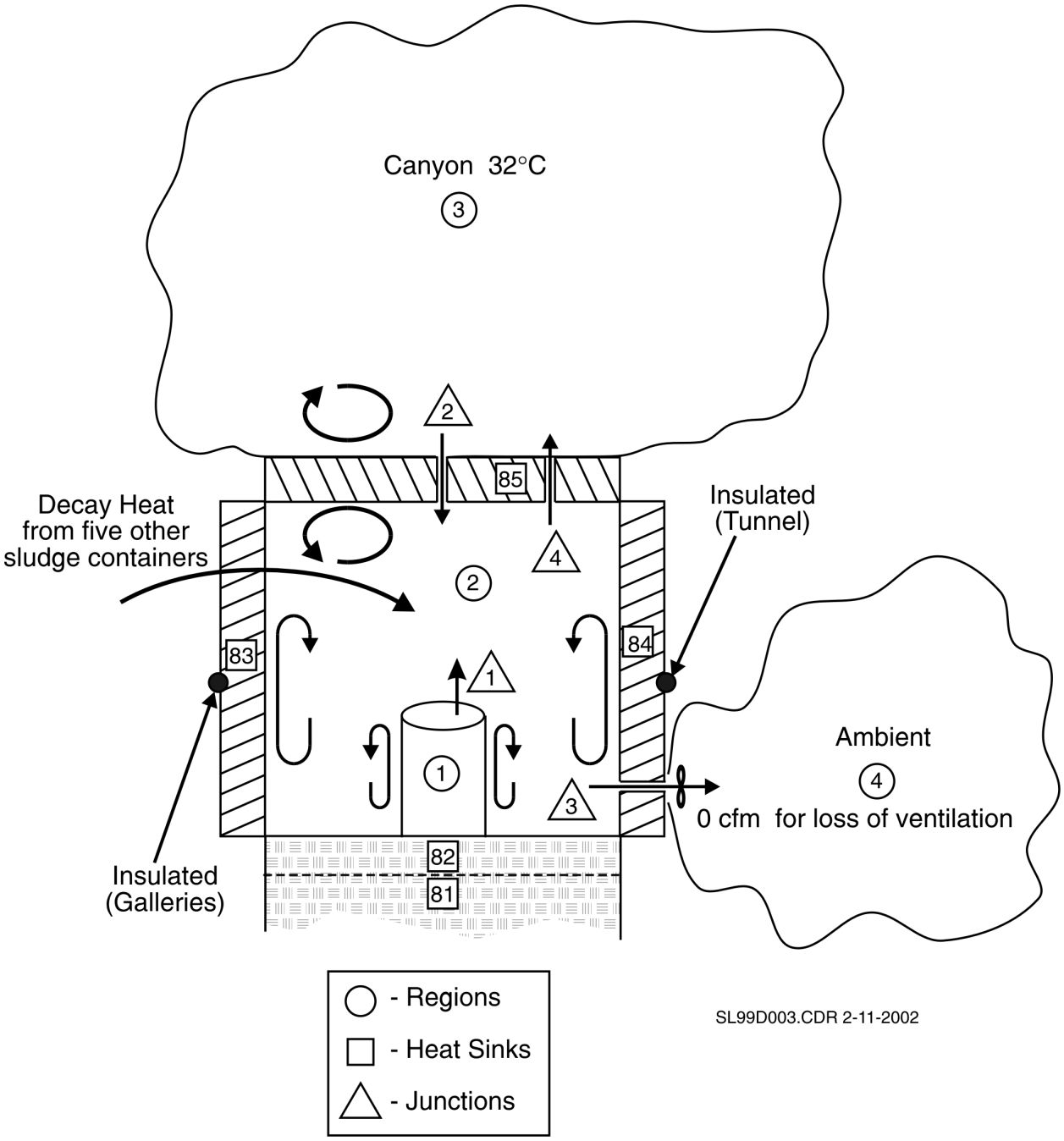

Figure 7. Nodalization and Processes for an LDC in a T Plant Cell.

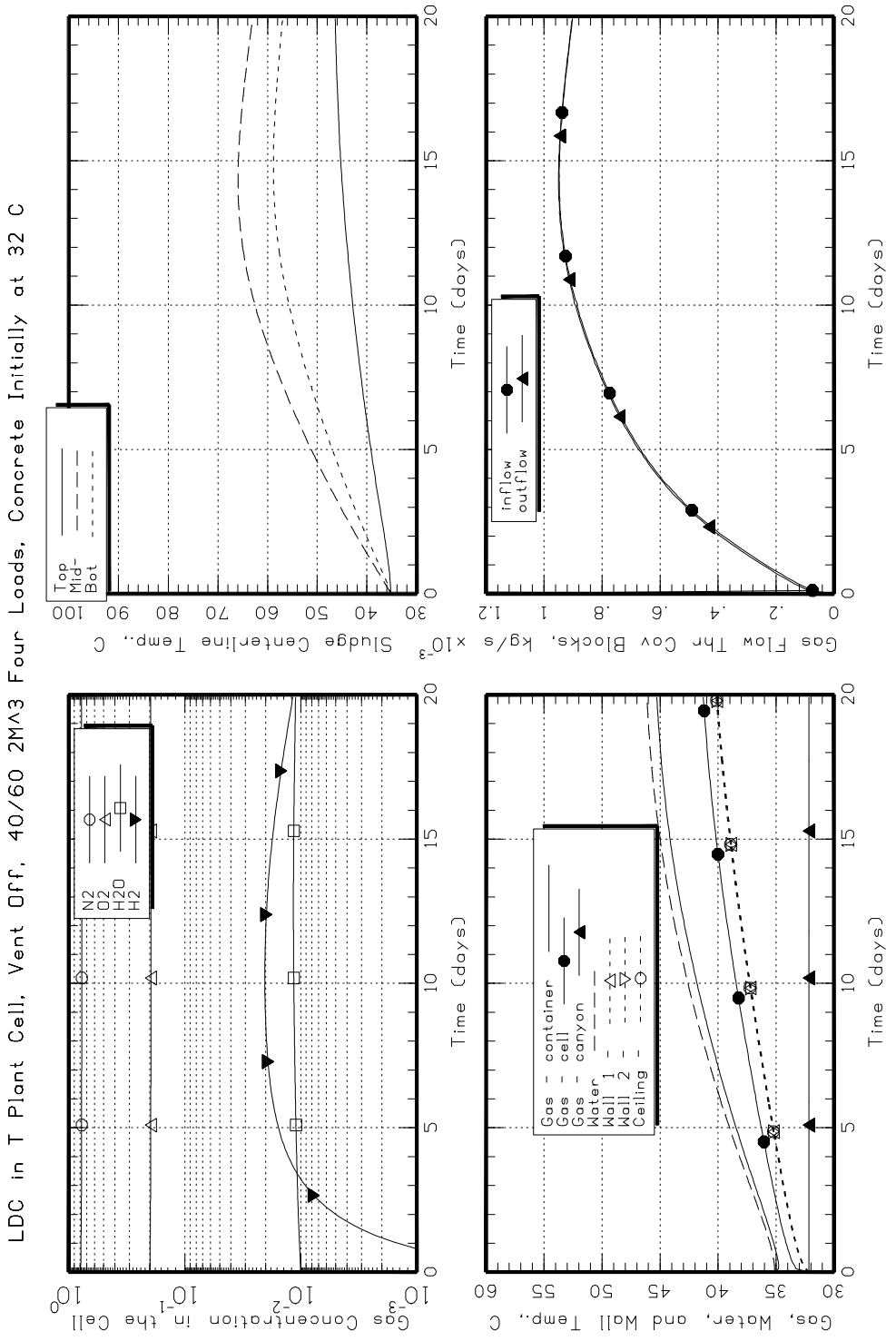

Figure 8. T Plant Fans Off Results: Cell Gas Composition, Sludge Temperature, and Cell Flow.

Conclusions from Integral Modeling

Integral models are used to utilize experimental information on uranium metal content, particle size, and limiting heat removal capability and to couple it with scenario-specific information for transportation and storage. Through evaluation of the detailed temperature profile in the sludge and associated chemical reaction rates, container design could be selected to prevent formation of flammable atmospheres during off-normal events. The computer code, HANSF, is generally applicable to other waste types and facilities.

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