

ASSESSMENT OF THE POTENTIAL FOR HYDROGEN GENERATION DURING GROUTING OPERATIONS IN C-REACTOR DISASSEMBLY BASIN

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**TITLE: Assessment of the Potential for Hydrogen Generation During Grouting Operations
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Assessment of the Potential for Hydrogen Generation During Grouting Operations in C-Reactor Disassembly Basin

Executive Summary

C-Reactor Disassembly Basin (DB) is being prepared for deactivation and decommissioning (D&D). D&D activities will consist primarily of immobilizing contaminated scrap components and structures in a grout-like formulation. The DB will be the first area of the C-Reactor Building that will be immobilized. The scrap components contain aluminum alloy materials. Any aluminum will corrode very rapidly when it comes in contact with the very alkaline grout ($\text{pH} > 13$), and as a result would produce hydrogen gas. To address this potential deflagration/explosion hazard, Savannah River National Laboratory (SRNL) reviewed and evaluated existing experimental and analytical studies of this issue to determine if any process constraints are necessary.

The risk of accumulation of a flammable mixture of hydrogen above the surface of the water during the injection of grout into the C-Reactor DB is low if the assessment of the aluminum surface area is reliable. Conservative calculations estimate that there is insufficient aluminum present in the basin areas to result in significant hydrogen accumulation in this local region. The minimum safety margin (or factor) on a 60% LFL criterion for a local region of the basin (i.e., Horizontal Tube Storage) was greater than 3. Calculations also demonstrated that a flammable situation in the vapor space above the basin is unlikely. Although these calculations are conservative, there are some measures that may be taken to further minimize the risk of developing a flammable condition during grouting operations.

1. Minimize the initial temperature of the water and grout as much as practical. Lower temperatures will mean lower hydrogen generation rates.
2. Ventilate the building above the basin rim as much as practical (e.g., leave doors open and operate fans) to further disperse hydrogen.
3. Minimize interruptions to the grout placement process as much as possible. Interruptions will result in higher water temperatures and hence higher hydrogen evolution rates.
4. Grout areas where the actual areal density ratios are the highest (e.g., Horizontal Tube Storage and Vertical Tube Storage) first. Thus, the areas that will produce the highest volume of hydrogen will have the maximum building volume in which to expand.

Introduction

The C-Reactor building is being prepared for deactivation and decommissioning (D&D). D&D activities will consist primarily of immobilizing contaminated components and structures in a grout-like formulation. The DB will be the first area of the C-Reactor Building that will be immobilized. The DB may be divided into six primary areas:

Horizontal Tube Storage (Final Storage or HTS), Vertical Tube Storage (VTS or Temporary Storage), Dry Cave, D&E Canal, Machine Basin (including the Monitor Basin) and the Transfer Station.

Currently contaminated scrap components from the reactor and basin are scattered across the basin floor in a random fashion (see Figure 1). Furthermore, the basin is filled with water to within 79 inches of the 0'-0" elevation. The current plan is to inject grout beneath the water in order to immobilize the scrap components and any sludge material on the basin floor.

The scrap components contain aluminum alloy materials [1]. Aluminum corrodes very rapidly when it comes in contact with the very alkaline grout ($\text{pH} > 13$), and as a result will produce hydrogen gas [2]. If the areal density ratio (i.e. surface area of aluminum to cross-sectional area of basin floor) exceeds a critical value, the volume percentage of hydrogen at the water surface could exceed the lower flammability limit (i.e., 4 vol.%). To address this potential deflagration/explosion hazard, Savannah River National Laboratory (SRNL) reviewed and evaluated existing experimental and analytical studies of this issue to determine if any process constraints are necessary.

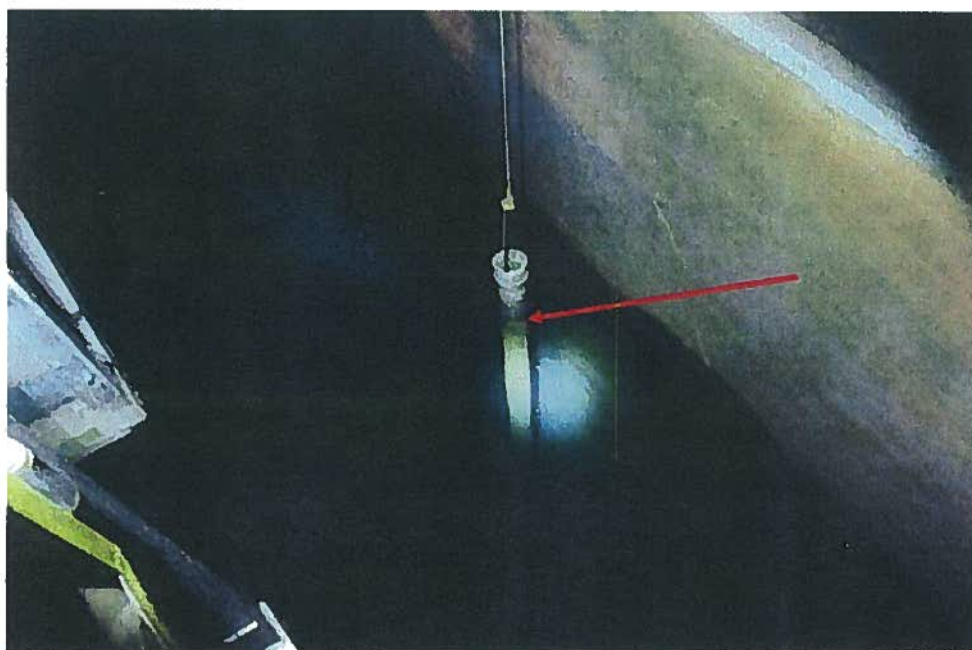


Figure 1. Universal Sleeve Housing (USH) located in the Vertical Tube Storage region of the basin.

Approach

Pacific Northwest National Laboratory (PNNL) developed a methodology for assessment of hydrogen generation during grouting operations at the K basins at the Hanford Site [3]. Likewise, similar analyses were performed for the R and P Reactor DBs at SRS [4, 5].

These documents were reviewed and applied to the situation for C-DB. The approach is as follows:

- 1) Aluminum corrodes upon exposure to the high pH grout.
- 2) Hydrogen is generated as a consequence of the corrosion reaction.
- 3) The gas rises to the surface of the water in the form of bubbles.
- 4) The bubbles will burst at the water surface releasing H_2 gas into the stagnant air layer.

This process was modeled by formulating a kinetic law for hydrogen production as a function of the grout temperature and combining it with a model for vertical turbulent diffusion of a light fluid (H_2) through a heavier miscible fluid medium (air). Vertical turbulent diffusion is a process analogous to molecular diffusion. However, the diffusion coefficient is several orders of magnitude larger than the molecular diffusion coefficient for the H_2 /air mixture, because vertical diffusion of the lighter gas is due to buoyancy rather than molecular motion. This model has been confirmed experimentally and has been shown to be effective for predicting diffusion layers that are broader than they are tall (i.e., similar to the basin floor [6]).

The assumptions used in the analysis were:

- The aluminum metal sources are situated on the bottom of the basin pool and are not covered by or contained within structures where hydrogen could accumulate.
- Once the hydrogen reaches the floor level of the basin (i.e., 0'-0" elevation), there is sufficient advection to disperse the hydrogen within the building superstructure.

Based on these assumptions the most likely location for hydrogen accumulation is in the region between the water surface and the 0'-0" elevation.

The first part of the model involved developing a kinetic law for the generation of hydrogen during the corrosion of aluminum. Laboratory tests were performed at PNNL to measure the hydrogen generation rate of non-corroded aluminum metal coupons immersed in both grout and in saturated $Ca(OH)_2$ solution. $Ca(OH)_2$ solution is formed when water is added to the grout. The key results from the tests were:

- The initial hydrogen generation rate of non-corroded aluminum metal in a grout mixture at 25 °C is 0.3 cm^3/min .
- The initial hydrogen generation rate of non-corroded aluminum metal in $Ca(OH)_2$ solution at 25 °C is 1.1 cm^3/min .
- The initial hydrogen generation rate of non-corroded aluminum metal in $Ca(OH)_2$ solution at 50 °C is 5.4 cm^3/min .
- The hydrogen generation rate of non-corroded aluminum metal in grout decreases to approximately 0.15 cm^3/min after 2 to 3 hours of exposure to the grout mixture at 25 °C. Likewise, the hydrogen generation rate of non-corroded aluminum metal in $Ca(OH)_2$ decreases to approximately 0.27 cm^3/min at 25 °C and approximately 0.18 cm^3/min at 50 °C. The decrease in corrosion rate is due to the

formation of a corrosion product (principally tricalcium aluminum hydroxide and hydrocalumite) layer on the surface of the aluminum metal.

The Arrhenius equation for the hydrogen generation rate was derived previously for grout and Ca(OH)_2 [4]. For grout, the volumetric hydrogen generation rate per unit area of aluminum (Q') is:

$$Q' = 55.68 \exp(-5339/T) \quad (1)$$

where T is the temperature in $^{\circ}\text{K}$. Similarly, for Ca(OH)_2 Q' is:

$$Q' = 262.1 \exp(-5339/T) \quad (2)$$

The following conservatisms and uncertainties were considered when applying the data to the C-DB situation.

- The experimentally measured hydrogen generation rate was determined on clean or non-corroded aluminum metal. The surface of the aluminum metal in the basins is corroded. The aluminum metal in the basins has been there for many years and the surface is protected by either a natural thin hydroxide film or a corrosion product layer. In either case, the rate of hydrogen generation from corrosion would be lower than that measured for the non-corroded aluminum metal coupons in the laboratory tests.
- The hydrogen generation rate due to aluminum metal corroding in grout is based on only one relevant gas generation test. However, four tests were conducted in a Ca(OH)_2 solution and the results were consistent based on chemical engineering fundamentals (i.e., mass transfer conditions in the grout are poorer than those in the Ca(OH)_2 solution). Additionally, the hydrogen gas generation rate for the aluminum in grout was also comparable to other values in the literature. Matsuo et al. measured a rate of $0.105 \text{ cm}^3/\text{min}$ for aluminum exposed to Portland cement at room temperature [7, 8].
- The hydrogen generation rate for aluminum metal in grout was performed at 27°C . Extrapolation of this result to hydrogen generation rates for higher grout temperatures was made using the test results from aluminum exposed to Ca(OH)_2 solution. However, the five-fold increase in hydrogen generation rate with a 30°C increase in the Ca(OH)_2 solution temperature is consistent with literature values for hydrogen generation in grout. Matsuo et al. observed a 3 fold increase in hydrogen generation rate with a 30°C increase in an inhibited grout mixture [9].

While the last two bullets do indicate that there is uncertainty in the experimental data due to the limited number of laboratory tests, it is unlikely that this is significant relative to other conservatisms in the analysis.

The mass transport equation was also derived previously [4]. The superficial velocity, u_0 , was expressed as:

$$u_0 = \beta^2 [gH (1 - (M_L/M_H) X_{LFL})^3]^{1/2} \quad (3)$$

where β is a proportionality constant equal to 1.64, g is the gravitational acceleration, H is the distance from the water-air interface to the basin's 0'-0" elevation, M_L is the molecular weight of hydrogen, M_H is the molecular weight of air, and X_{LFL} is the volume fraction of hydrogen at the lower flammability limit (LFL).

The following conservatisms and uncertainties were considered when applying the diffusion model to C-basin.

- 1) The analysis does not account for dissipation of hydrogen between the surface of the water and the 0'-0" elevation of the basin due to advection. Accounting for this phenomenon would minimize the accumulation of hydrogen in this region.
- 2) The analysis assumed that the hydrogen bubble plume does not expand laterally from the aluminum metal source. In reality, local concentrated sources of aluminum metal will produce a bubble plume which expands laterally as it rises through the water. Thus, the potential for a local deflagration/explosion is overestimated in this analysis.

The incipient flammability condition occurs when the gas generation rate due to corrosion equals the flux of hydrogen through air. The critical condition occurs at the water-air interface when the hydrogen gas concentration exceeds the LFL. The LFL for hydrogen in air is 4% by volume. To assess safety margins, the calculations were also performed with a criterion of 60% of the LFL. This value is utilized for safety class operations that involved the handling of radioactive storage vessels [10].

The critical areal density ratio can be derived by equating the kinetic law for hydrogen generation rate to the vertical turbulent diffusion rate of hydrogen when the hydrogen concentration at the surface of the water is the LFL. This derivation was also performed previously [4]. The critical areal density ratio is defined as:

$$(A_{Al}/A_{floor})_c = u_0/Q' \quad (1)$$

where A_{Al} is the surface area of the aluminum, A_{floor} is the cross-sectional surface area hydrogen will diffuse through, u_0 is the superficial velocity of the hydrogen gas as it diffuses toward the 0'-0" elevation of the basin, and Q' is generation rate of hydrogen due to corrosion of aluminum. The actual areal density ratio will be compared to the calculated critical ratio. If the actual ratio is greater than the critical calculated ratio, there is risk that the concentration of hydrogen gas at the surface of the water in the basin is greater than the lower flammability limit (LFL). Further precautions would be necessary if this condition exists. Two cases were considered, 1) aluminum exposed to grout only and 2) aluminum exposed to $Ca(OH)_2$ solution. The latter case is considered in grouting operations where there are interruptions for operational or other reasons. In this situation the water is assumed to become saturated with $Ca(OH)_2$.

Figure 2 shows a plot of the critical areal density ratio as a function of temperature for aluminum exposed to grout. The ratio decreases with temperature as expected due to the increase in corrosion rate, and hence hydrogen generation rate, with temperature. The plot also shows the critical areal density ratio for both 100% and 60% LFL.

Figure 3 shows a similar plot for aluminum exposed to $\text{Ca}(\text{OH})_2$. The critical areal density ratios are lower than those for the grout, which reflects the higher aluminum corrosion rate. Again the plot also shows the critical areal density ratio for both 100% and 60% LFL.

Heat is added to the system during the grout curing process through the heat of hydration. The heat of hydration is the heat evolved due to chemical reactions with water and is dependent upon the constituents present in the cement mix. Experimental data from the literature suggests that the maximum temperature does not occur until 10-20 hours after the curing process begins (See Figure 4 [9]). During the first 3 hours, when hydrogen generation is at a maximum, the temperature is expected to be less than 50 °C [9]. Since the corrosion reaction decreases significantly after 3 hours due to the formation of corrosion products on the aluminum surface, it is the first 3 hours that are the most critical from the standpoint of hydrogen evolution. Thus, for this evaluation it will be assumed that the maximum temperature of the grout is 50 °C.

One concern is that the grout placement will be interrupted for operational or other reasons. If this is the case, the temperature of the water above the concrete will begin to increase due to the heat of hydration. An energy balance was performed to estimate the maximum temperature rise of the $\text{Ca}(\text{OH})_2$ solution. The conservatisms built into the analysis include:

- The energy balance assumed that the grout and basin water were a closed system and there was no thermal losses to the basin structure or to the air above the basin. Only heat conduction from the grout to the basin water was assumed, completely ignoring heat loss via natural convection.
- The quantity of heat generated by the heat of hydration used for the PNNL thermal analysis was for a cumulative 7 day period. This input is extremely conservative given that the critical time period of interest is the first 3 hours after exposure to $\text{Ca}(\text{OH})_2$. For the C-DB the heat released during the first day was assumed to be 25% of the heat released during the 7 days. This assumption takes into account that the heat generation rate is high initially; however, heat generation is not significant to the hydrogen generation rate after the first 3 hours. Therefore, it is less than the cumulative heat generated that is assumed in the PNNL analysis.

The calculations are shown in Appendix 1. The calculation was performed for various grout pour heights, in order to simulate different interruption scenarios. The temperature rise also depends on the depth of the water. The depth of the water in the Horizontal Tube Storage (HTS) area was approximately 12 feet, while in the remaining areas the depth is approximately 23 feet. Therefore, for HTS, the maximum calculated

temperature was 62 °C, while for the other areas the maximum temperature was 40°C. This evaluation is conservative as it assumes that operations were interrupted near the conclusion of the initial five foot lift.

Determination of Cross-Sectional Area of the Basin and Actual Aluminum Surface Area

The assessment was performed to ensure that hydrogen does not accumulate within a local region of the DB . As mentioned previously, the DB is divided into five primary regions. Drawings of these areas were studied to determine the cross-sectional area that the hydrogen will diffuse through during the grouting operation [11]. The localized area where most of the aluminum components were located was determined in consultation with D&D Engineering (see Appendix 2). The calculations are shown in the Appendix 1, and are summarized in Table 1.

Table 1. Cross-Sectional Area of the Disassembly Basin Regions and Aluminum Component Surface Area

Region of the Basin	Cross-Sectional Area (ft²)	Aluminum Surface Area (ft²)
D&E Canal	256	51
Horizontal Tube Storage	2180	9202
Vertical Tube Storage	585	3114
Machine Basin	1989	77
Dry Cave	735	394

D&D Engineering personnel performed a field walk-down to determine the configuration of the aluminum components [1, 12]. Aluminum components that were identified include: universal sleeve housing (USH), semi-permanent sleeves, safety rod thimble, target bundle, harp, and aluminum scrap. D&D Engineering performed calculations to estimate the total surface area of aluminum in each region of the basin [12]. These values are summarized in Table 2. The assessment was performed under the assumption that the aluminum components were uniformly distributed in a local region of the basin floor.

Determination of Minimum Ventilation Rate

If the building was not properly ventilated, conceivably hydrogen could accumulate in the building vapor space at concentrations greater than the LFL. To evaluate this risk, the building was modeled as continuous stirred tank reactor (CSTR). The derivation of the model equations are shown in Appendix 1. The model predicts the accumulation of hydrogen as a function of time and assumed ventilation rate.

Two bounding cases were evaluated. The first case assumes that the aluminum is exposed to grout at a temperature of 50 °C, while the second case assumes that the aluminum is exposed to Ca(OH)₂ at 62 °C. Two criteria were used to assess the ventilation rates.

- 1) The ventilation rate needed to keep the hydrogen concentration less than 60% LFL and LFL for the first day was determined. Given that the corrosion rate decreases significantly after the first day, most of the hydrogen will accumulate during this time.
- 2) The time necessary for the hydrogen concentration to exceed 60% LFL and LFL was calculated. For this analysis the gas generation rate will be assumed to be constant and equal to the time averaged, one day hydrogen generation rate. This assumption is conservative for two reasons. First, the generation rate is known to decrease significantly due to passivation of the corrosion reaction. Secondly, the analysis will assume an infinite supply of aluminum in the basin, which clearly is not the case. Nevertheless, this criterion should show the benefits of providing ventilation during the operations.

Results

Figure 5 summarizes the flammability assessment for each of the basin regions for the condition where the aluminum is exposed to grout. The grout was assumed to be at the maximum temperature of 50 °C. The highest actual areal density ratios were observed for the Horizontal Tube Storage, 4.22, and the Vertical Tube Storage, 5.32. The critical areal density ratio for the 60% and 100% criteria is 116 and 249, respectively. The margins of safety for Horizontal Tube Storage (HTS) and Vertical Tube Storage (VTS) are a factor of 27 and 22, for the 60% criterion, respectively. This margin of safety was considered adequate (i.e., one to two orders of magnitude) when considering the usage of calcium aluminate sulfate grout for the P-reactor grouting operations [13]. At the 100% criterion these margins of safety increase to 59 for the HTS region and 47 for the VTS region.

For perspective, in order to achieve the critical areal density ratio for the 60% criterion, the HTS region would need to contain an additional 3800 USHs. This value more than doubles to an additional 8150 USHs if the 100% criterion is utilized. For the VTS area, the critical areal density ratio for the 60% criterion could be achieved if there are more than 980 additional USHs present; an additional 2170 USHs would need to be present for the 100% criterion to be exceeded. Therefore, if the D&D Engineering is confident in the results of the field observations [1, 12], the development of a flammable situation in the basin is unlikely.

Figure 6 summarizes the flammability assessment for each of the basin regions, except HTS, for the condition where the aluminum is exposed to Ca(OH)_2 . The Ca(OH)_2 was assumed to be at a temperature of 40 °C. The critical areal density for the 60% and 100% criteria for these regions is 42 and 90, respectively. The margin of safety for VTS is a factor of 8 for the 60% criterion. Although this margin is less than for the grouted condition, this margin of safety is also adequate [13]. At the 100% criterion, the safety factor for this region is 17.

In order to achieve the critical areal density ratio for the 60% criterion, the Vertical Tube Storage region would need to contain an additional 325 USHs; for the 100% criterion an

additional 755 USHs would need to be present. This result indicates that the margin for safety is less if the operations were to be suspended before the five foot lift was completed. However, if D&D Engineering is confident in the results of the field observations [1, 12], the development of a flammable situation in the basin is unlikely.

Due to the shallower depth of HTS (i.e., 19 ft), there is less water available to heat. Therefore, the maximum temperature for the $\text{Ca}(\text{OH})_2$ during a five foot lift was calculated to be 62 °C. The critical areal density for the 60% and 100% criteria for these regions is 14 and 29, respectively. The margins of safety for HTS are a factor of 3.3 for the 60% criterion and 6.9 for the 100% criterion. These safety factors are less than an order of magnitude, however, would be equivalent to limiting maximum allowable hydrogen concentration to less than 15% of the LFL. A review of the safety requirements for the Tank Farm Facility indicates that this application would be analogous to a very slow generating tank [14]; that is, a tank where the hydrogen concentration will not achieve the lower flammability limit at steady state. Special controls are required in very slow generating tanks only if the steady state value exceeds 60% LFL. Thus, the calculations indicate that there is a sufficient margin of safety.

In order to achieve the critical areal density ratio for the 60% criterion, HTS would need to contain an additional 460 USHs; for the 100% criterion an additional 800 USHs would need to be present. This result indicates that there that the margin for safety is less if the operations were to be suspended before the five foot lift was completed. However, if D&D Engineering is confident in the results of the field observations [1, 12], the development of a flammable situation in the basin is unlikely.

Figures 7 and 8 summarize the results for the assessment of the effect of ventilation on the accumulation of hydrogen within the building above the basin area. For the case where the aluminum is exposed to grout at 50 °C, the accumulation of hydrogen for three ventilation rates is shown in Figure 7. After Day 1 there is little effect of ventilation rate on the hydrogen accumulation. In all three cases, the concentration of hydrogen was on the order of 0.045 vol.%, which is approximately two orders of magnitude less than the LFL and a factor of 50 less than the 60% LFL criterion. These concentrations were observed even when the ventilation rate was on the order of 0.1 cubic foot/min.

The benefit of ventilation can be seen if the operations are extended for more than a day. Figure 7 shows that for a ventilation rate of 5 cubic feet/min, the hydrogen concentration will barely exceed 60% LFL and will never achieve the LFL. At 0.1 cubic foot/min the hydrogen concentration above the basin was calculated to exceed 60% LFL after 52 days and LFL after 87 days. As mentioned before, these concentrations are conservative as they do not account for the decrease in hydrogen generation due to passivation of the aluminum. On the other hand, the uncertainties in the surface area of aluminum exposed also factor into this assessment as they did for the assessment of local accumulation of hydrogen. However, if D&D Engineering is confident in the results of the field observations [1, 12], the development of a flammable situation in the vapor space above the basin is unlikely.

For the case where the aluminum is exposed to $\text{Ca}(\text{OH})_2$ at 62 °C, the accumulation of hydrogen for four ventilation rates is shown in Figure 8. After Day 1, little effect of ventilation rate on the hydrogen accumulation was observed. In all four cases, the concentration of hydrogen was on the order of 0.35 vol.%, which is more than an order of magnitude less than the LFL and a factor of 8 less than the 60% LFL criterion. These concentrations were observed even when the ventilation rate was on the order of 0.1 cubic foot/min.

Figure 8 shows that for a ventilation rate of 56 cubic feet/min, the hydrogen concentration was calculated to never exceed 60% LFL, while at 33 cubic feet/min the hydrogen concentration does not achieve the LFL. At 0.1 cubic foot/min the hydrogen concentration is calculated to exceed 60% LFL after 6.8 days and LFL after 11.3 days. As mentioned before, these concentrations are conservative as they do not account for the decrease in hydrogen generation due to passivation of the aluminum. On the other hand the uncertainties in the surface area of aluminum exposed also factor into this assessment as they did for the assessment of local accumulation of hydrogen. However, if D&D Engineering is confident in the results of the field observations [1, 12], the development of a flammable situation in the vapor space above the basin is unlikely.

Conclusions and Recommendations

The risk of accumulation of a flammable mixture of hydrogen above the surface of the water during the injection of grout into the C-Reactor DB is low if the assessment of the aluminum surface area is reliable. Conservative calculations estimate that there is insufficient aluminum present in the basin areas to result in significant hydrogen accumulation in this local region. The minimum safety margin (or factor) on a 60% LFL criterion for a local region of the basin (i.e., Horizontal Tube Storage) was greater than 3. Calculations also demonstrated that a flammable situation in the vapor space above the basin is unlikely. Although these calculations are conservative, there are some measures that may be taken to further minimize the risk of developing a flammable condition during grouting operations.

1. Minimize the initial temperature of the water and grout as much as practical. Lower temperatures will mean lower hydrogen generation rates.
2. Ventilate the building above the basin rim as much as practical (e.g., leave doors open and operate fans) to further disperse hydrogen.
3. Minimize interruptions to the grout placement process as much as possible. Interruptions will result in higher water temperatures and hence higher hydrogen evolution rates.
4. Grout areas where the actual areal density ratios are the highest (e.g., Horizontal Tube Storage and Vertical Tube Storage) first. Thus, the areas that will produce the highest volume of hydrogen will have the maximum building volume in which to expand.

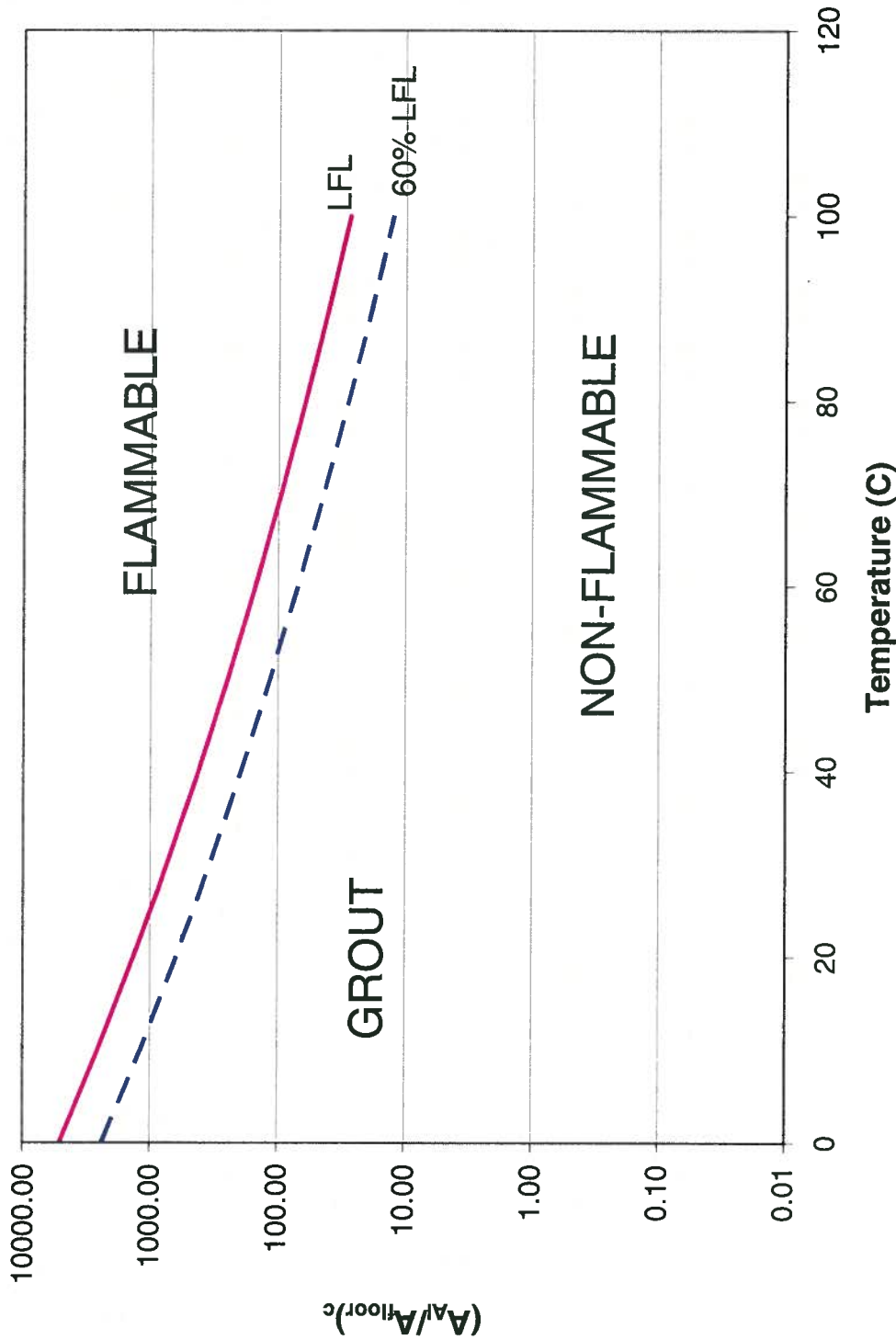


Figure 2. Aluminum Areal Density Ratio for Flammable Condition in Grout.

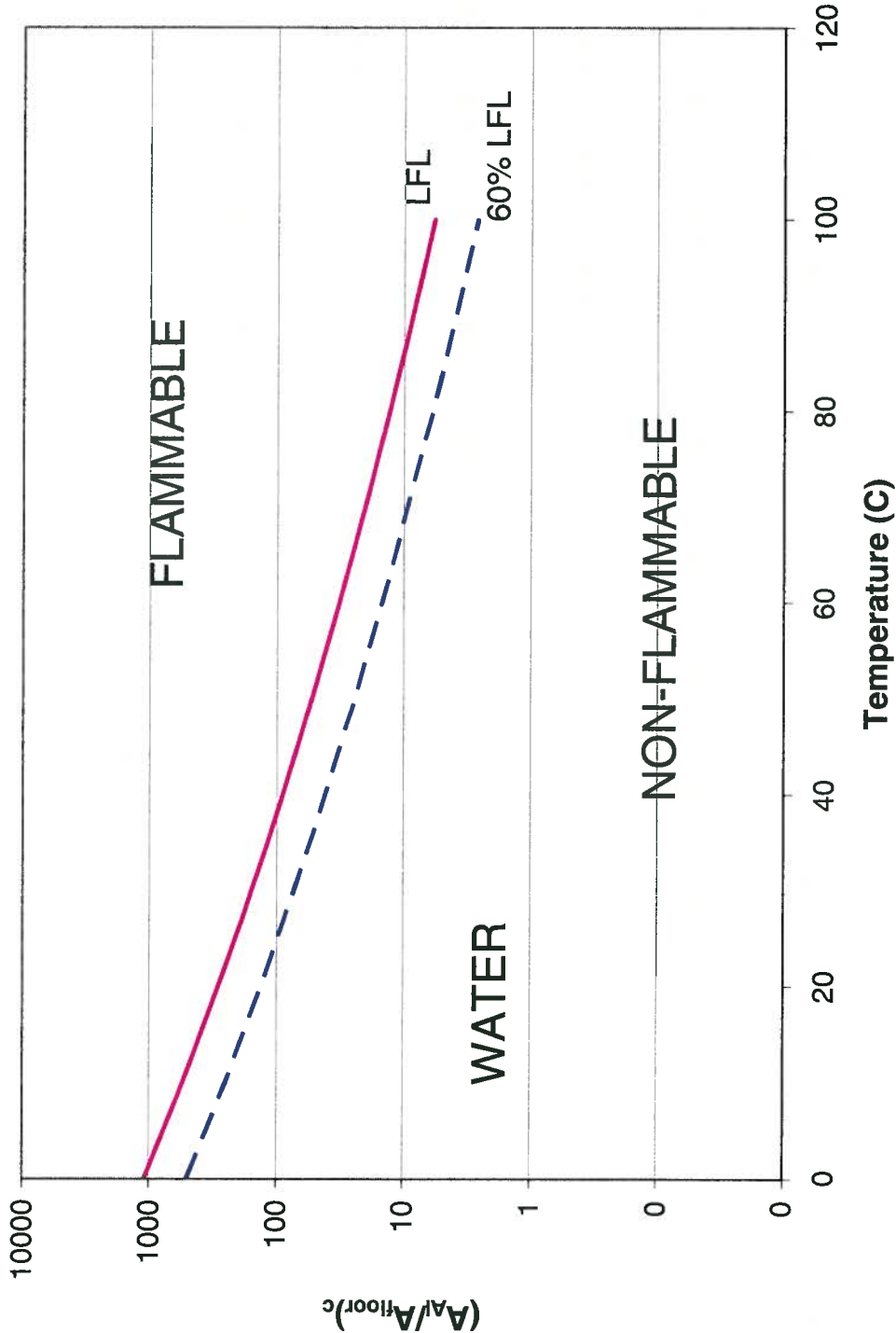
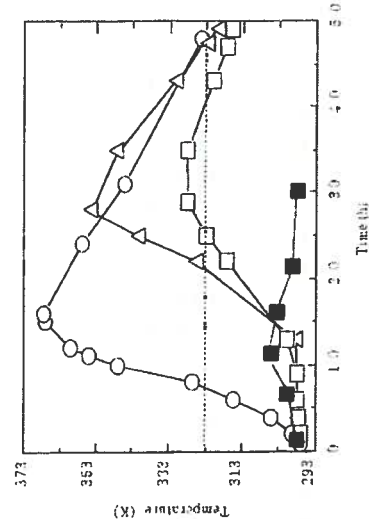


Figure 3. Aluminum Areal Density Ratio for Flammable Condition in $Ca(OH)_2$.







Case	Notation	Compositions of Mortar						Volume of Mortar Form (m ³)
		Compositions of Dry Mortar (wt%)				Water/OPC + BFS (wt%)		
		Case		OPC	BFS			
1		1	100.00	0	0	0	36.60	2 × 10 ⁻³
2		2	45.00	55.00	0	0		
3		3	24.75	30.25	45.00	0		
4		4	100.00	0	0	0		

Figure 4. Time dependence of temperature at the center of a mortar form for various cementitious materials. [7]

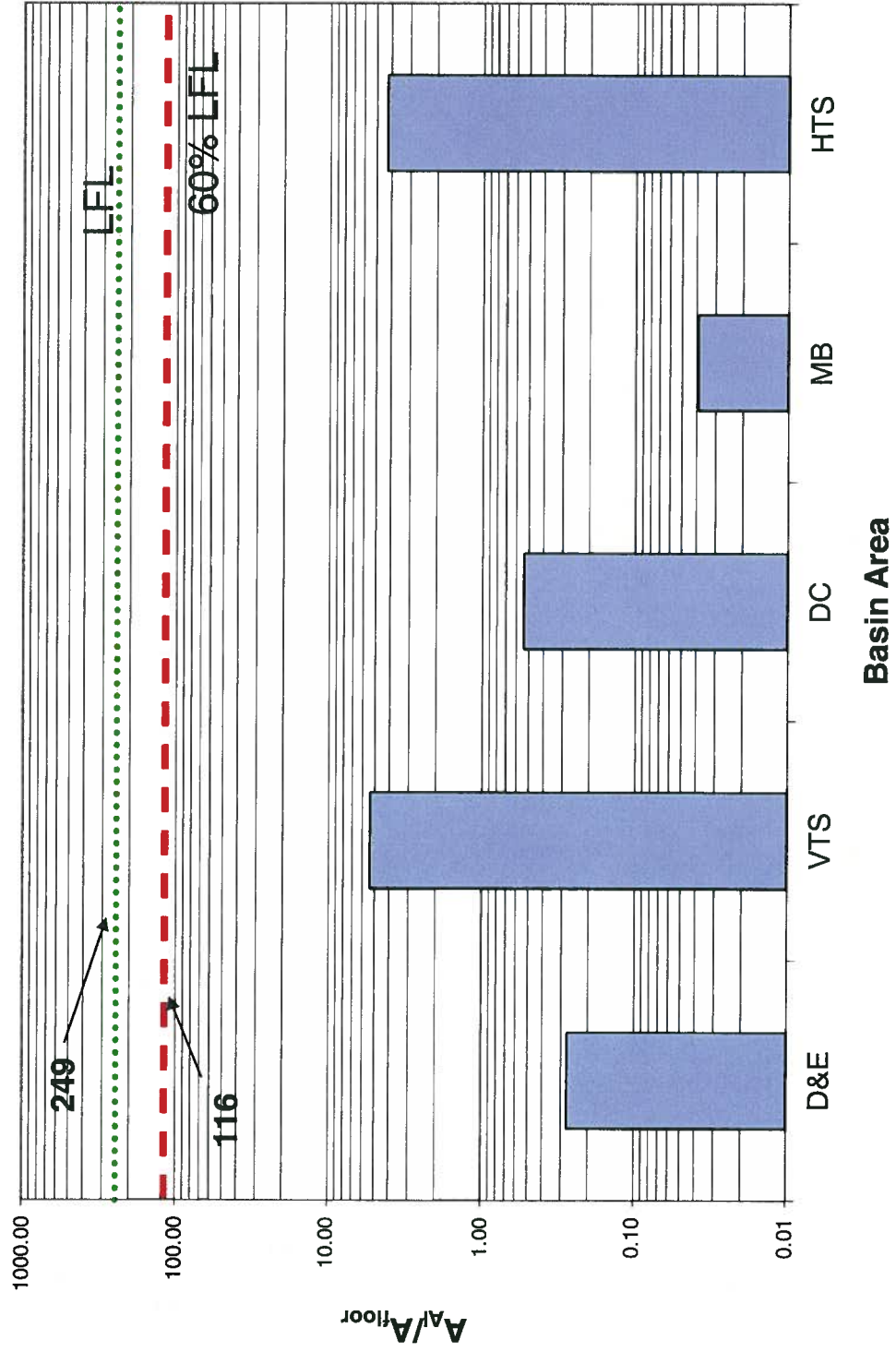


Figure 5. Assessment of Flammability Condition for Grout at 50 °C.

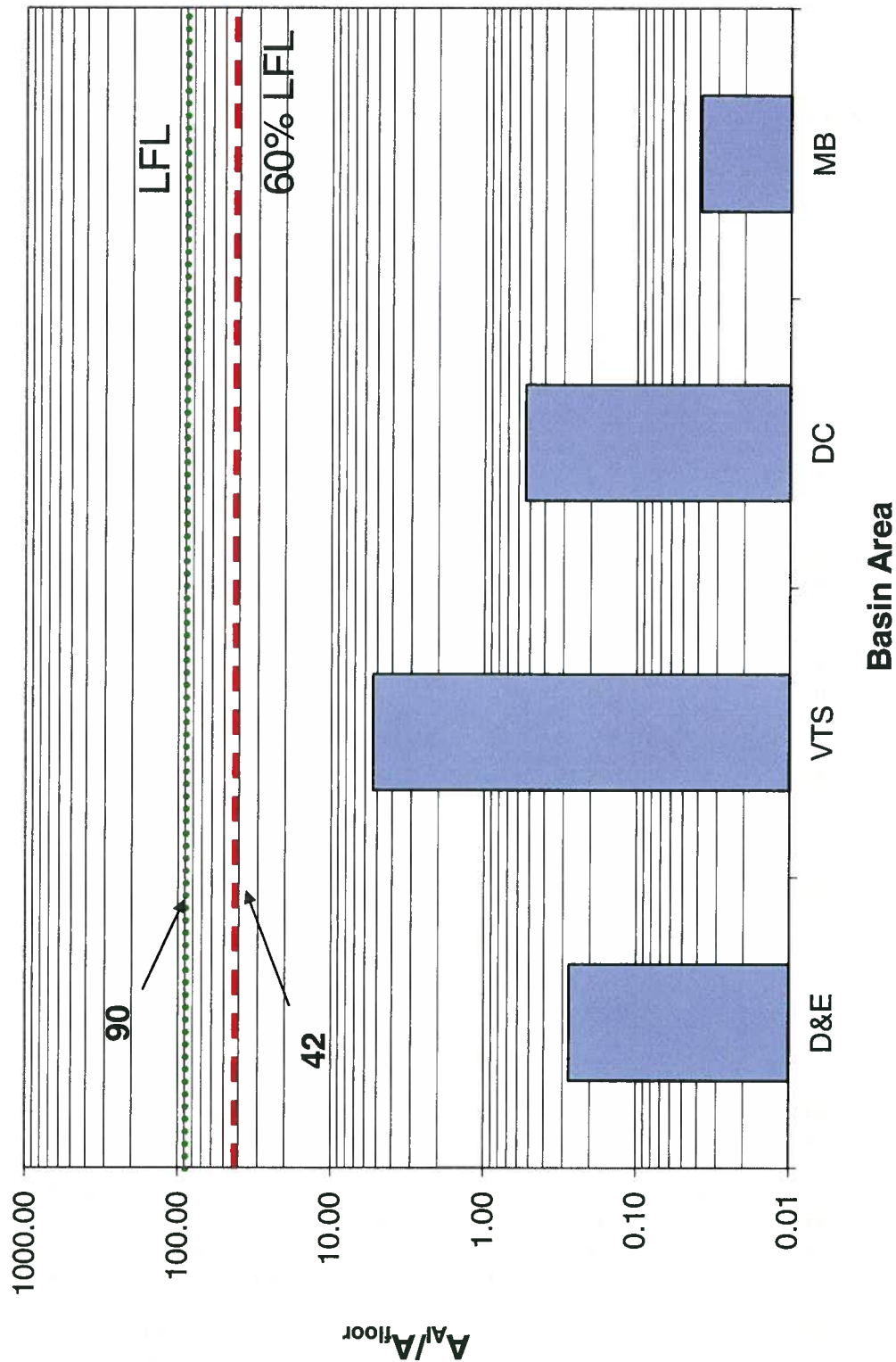


Figure 6. Assessment of Flammability Condition for $Ca(OH)_2$ at 40 °C.

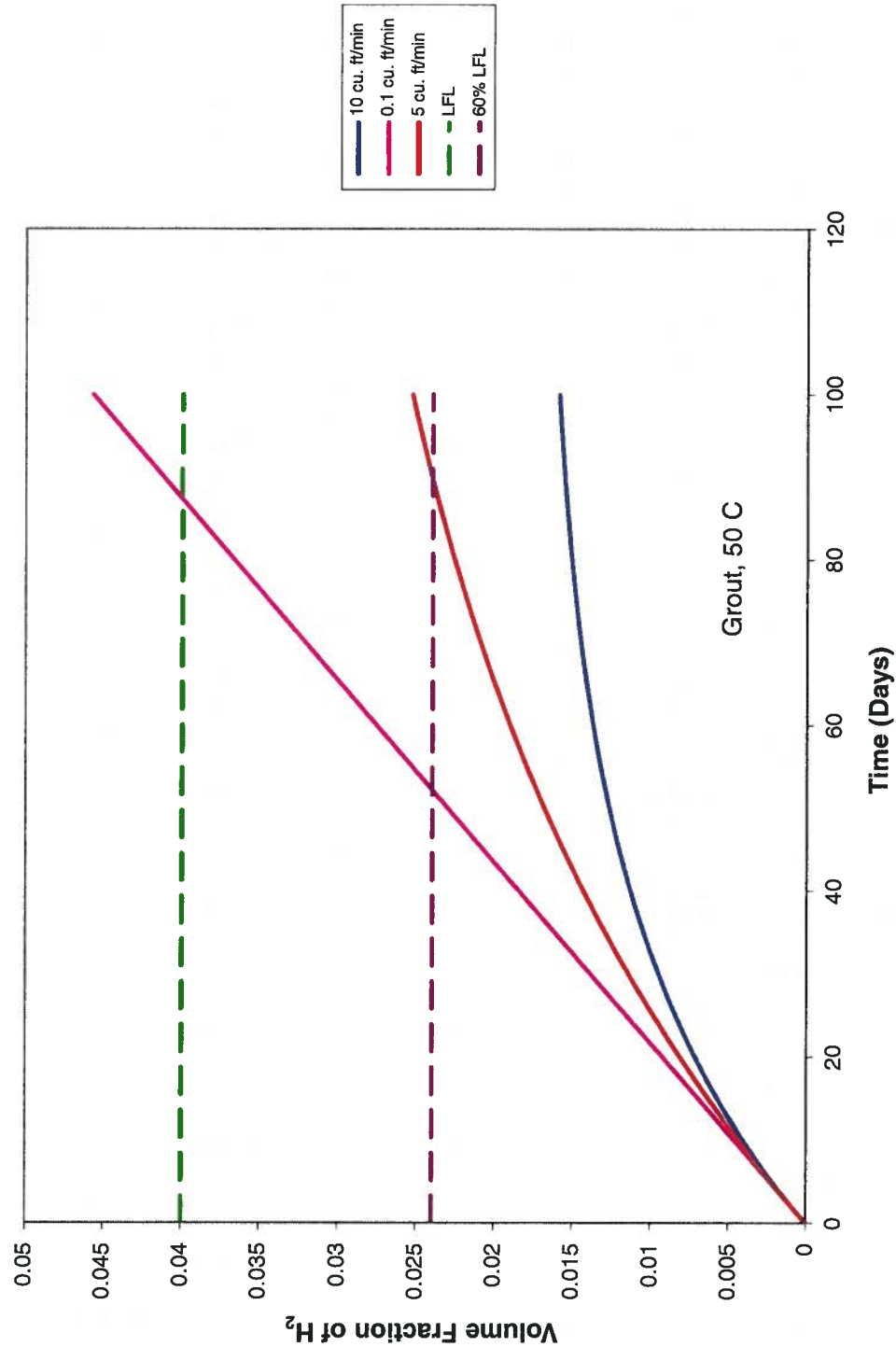


Figure 7. Assessment of Hydrogen Accumulation as a Function of Building Ventilation for Aluminum Exposed to Grout at 50 °C.

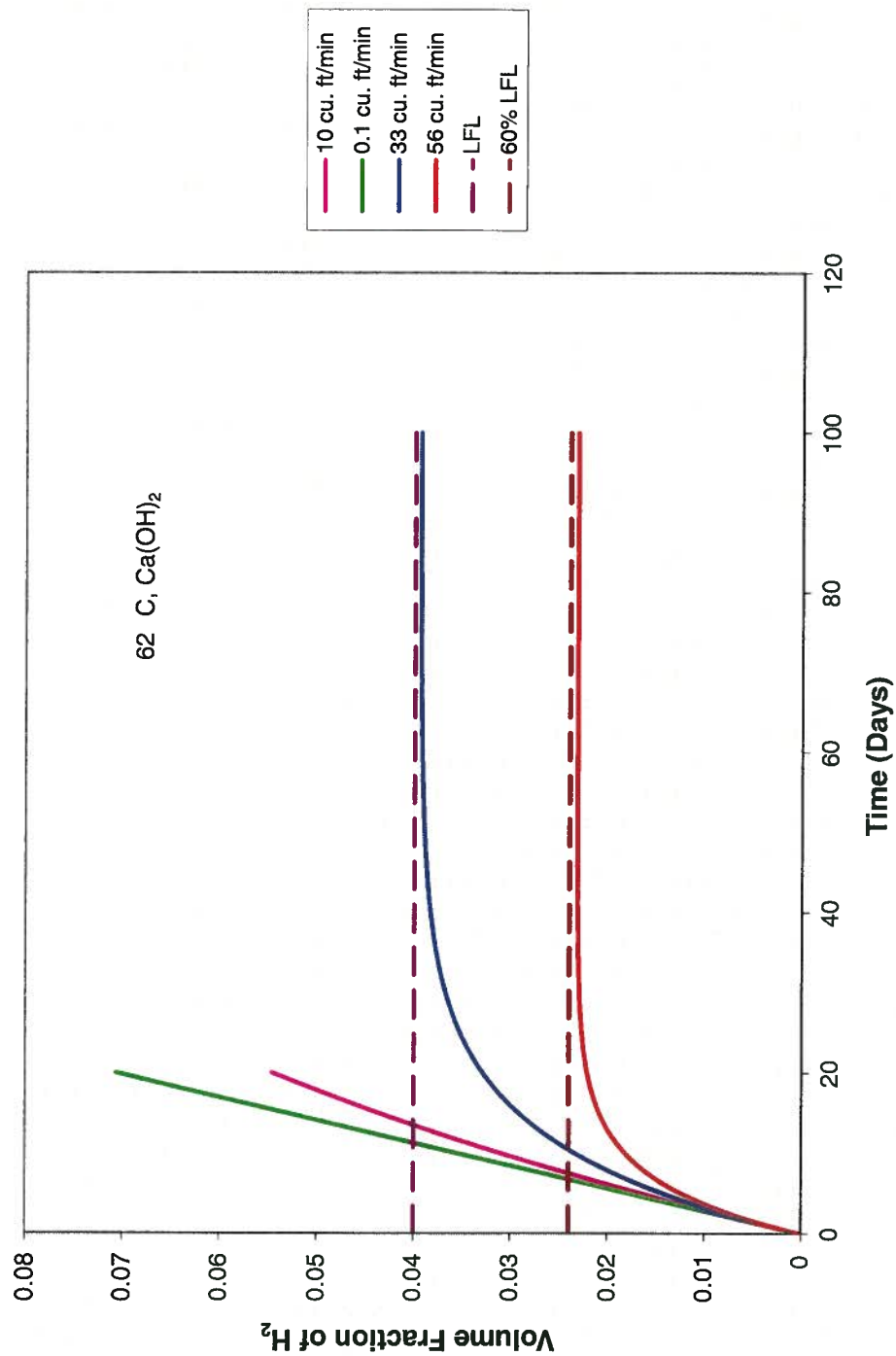


Figure 8. Assessment of Hydrogen Accumulation as a Function of Building Ventilation for Aluminum Exposed to Ca(OH)_2 at 62 °C.

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5. B. J. Wiersma, "Assessment of the Potential for Hydrogen Generation During Grouting Operations in P-reactor Disassembly Basin" SRNL-STI-2009-00364, June, 2009.
6. M. Epstein and J. P. Burelbach, "Transient Vertical Mixing by Natural Convection in a Wide Layer", Int. J. Heat Mass Transfer, Vol. 43, pp. 321-325.
7. T. Matsuo, T. Nishi, and M. Matsuda, "LiNO₃ Addition to Prevent Hydrogen Gas Generation from Cement-Solidified Aluminum Wastes", J. Nuclear Science Technology, Vol. 32, No. 9, pp. 912-920, 1995.
8. T. Matsuo, et al., "LiNO₃ Effect on Corrosion Prevention of Aluminum with Complex Shapes", J. Nuclear Science and Technology, Vol. 34, No. 8, pp. 823-828, 1997.
9. T. Matsuo, et al., "Influence of Increased Temperature from Cement Hydration on Aluminum Corrosion Prevention when LiNO₃ is Added to the Cement", Nuclear Technology, Vol. 125, pp. 332-336, March 1999.
10. B. J. Wiersma, "Assessment of the Potential for Hydrogen Generation During Grouting Operations in the R- and P-Reactor Vessels" SRNL-STI-2009-00639, Rev. 2, June, 2010.
11. C-Basin Drawings

W134454	W134417	W134473	W134486
---------	---------	---------	---------
12. W. B. Griffin, "Revised Calculation of the Surface Area of Aluminum Components and Material in 105-C Disassembly Basin", SDD-2011-00130, October 4, 2011. Calculation G-CLC-C-00022, Revision 1 is attached to memo.
13. B. J. Wiersma, "Compatibility of Stainless Steel and Aluminum Reactor Components with the Calcium Aluminate Sulfate Mix Design Utilized for Grouting of the P-Reactor Vessel", SRNL-STI-2010-00600, October, 2010.
14. B. C. Landeene, "CSTF Flammability Control Program", WSRC-TR-2003-00087, Rev. 17, December 2010.

APPENDIX 1

Title of Project C Reactor Disassembly Basin H₂

Project No. _____

Subject _____

Works _____

Computer BJWiersmaDate 10/25/11Sheet No. 1

Background

- Aluminum component debris is located near the bottom of the C reactor disassembly basin. There are five local areas in the basin: Dry caves, Horizontal Tube Storage, Vertical Tube Storage, Machine Basin, and D&E Canal.
- Location and number of components were determined during a field walkdown by D&D Engineering personnel (Ref. 1)
- Surface area of immersed aluminum were calculated by D&D Engineering personnel (Ref. 2).
- The water level in the basin is 79" below floor level. During grouting operations the water will evaporate such that the water level remain constant.
- The depth of the Horizontal Tube Storage area is 17 feet. The depth of all remaining areas is 30ft, with the exception of small isolated areas that have a depth of 50 feet
- Aluminum materials are located on the floor of the basin.
- The grout will be poured in 5 foot lifts. Most of the components will be covered by the first lift.

-
- 1) W.B. Griffin, et. al., "Building 105-C Disassembly Basin (DB) Engineering Guidance for Scrap Relocation", SDD-2011-00058, Rev. 0, May 17, 2011.
 - 2) W.B. Griffin, "Revised calculation of the Surface Area of Aluminum Components and Material in 105-C Disassembly Basin", SDD-2011-00130, October, 2011.

Title of Project C-Reactor Disassembly Basin H₂

Project No. _____

Subject _____

Works _____

Computer BJ WiersmaDate 10/25/11Sheet No. 2

Background (cont.)

- Corrosion will occur in both the water and grout. The pH of the water will increase with time due to exposure to the grout. If a lift is suspended, these calculations assume that the aluminum is exposed to $\text{Ca}(\text{OH})_2$
 - Calculation determines whether or not hydrogen accumulates at the surface of the water during grouting operations.
 - 60% LFL criterion will be used to determine margin on safety (ref.4)
- Model

Determination of Superficial Velocity (u_0) (ref.3)

See spreadsheet C-basin H₂ generation.xls, worksheet VTD calcs

$$u_0 = \beta^2 [gH(1 - (M_L/M_H) X_{LFL}^3)]^{1/2}$$

β = proportionality constant = 0.164

g = gravity = 9.8 m/s²

H = distance from water to floor = 79 inches = 2.0 meters

M_L = Molecular weight of H₂ = 2 g/mole

M_H = Molecular weight of air = 29 g/mole

X_{LFL} = 0.04 for calculations assume 60% LFL will provide safety margin $\therefore X_{LFL} = 0.024$ (ref.4)

$$u_0 = 4.27 \times 10^{-4} \text{ m}^3/\text{m}^2\text{-s}$$

Reference

- B.J. Wiersma, "Assessment of the Potential for Hydrogen Generation During Grouting Operations in R-Reactor Disassembly Basin", SRNL-STI-2009-00278, April 2009.

Title of Project C Reactor Disassembly Basin H₂Project No. p. A1-4Subject Works Computer BTWDate 10/25/11Sheet No. 4

Determination of Critical Areal Density Ratio

$$\left(\frac{A_{AI}}{A_{floor}} \right)_c = \frac{u_0}{\dot{Q}_{H_2}}$$

Determine A_{AI}/A_{floor} as a function of temperatureSee "C-basin H₂ Generation" spreadsheet, worksheet VTD calcs

Areal Density Ratio for Grout

T(°C)	$(A_{AI}/A_{floor})_c$
0	2393
10	1199
20	630
30	345
35	259
40	197
50	116
60	71
70	44
80	28
90	19
100	13

Areal Density Ratio for Ca(OH)₂

T(°C)	$(A_{AI}/A_{floor})_c$
0	508
10	255
20	134
30	73
35	55
40	42
50	25
60	15
70	9
80	6
90	4
100	3

Note: $X_{LFL} = 0.024$ or 60% LF

Determination of Amount of Aluminum

Summary of Reference 2

Area	A_{AI} (ft ²)
Horizontal Tube Storage	9202
Machine Basin	77
Dry Cave	394
Dry Cave - Center Pen	394
Vertical Tube Storage	3114
D & E Canal	70

Title of Project C-Reactor Disassembly Basin H₂

Project No. _____

Subject _____

Works _____

Computer BJ WiersmaDate 10/25/11Sheet No. 3

Corrosion Rate / Hydrogen Generation Rate (see ref.3)

For Grout

$$Q'_{H_2} = 55.68 \exp(-5339/T) \quad T = \text{Temperature in } ^\circ K$$

For $Ca(OH)_2$

$$Q'_{H_2} = 262.1 \exp(-5339/T)$$

Calculate Q'_{H_2} as a function of T . Shown in spreadsheet "C-basin H₂ generation" on worksheets "Mol Diffusion Calcs" and "VTD calcs"

H₂ Generation for grout

$T (^{\circ}K)$	$Q'_{H_2} (m^3/m^2 \cdot s) \times 10^7$
273	1.79
283	3.57
293	6.79
303	12.4
308	16.5
313	21.7
323	36.9
333	60.6
343	96.7
353	150
363	228
373	338

H₂ Generation for $Ca(OH)_2$

$T (^{\circ}K)$	$Q'_{H_2} (m^3/m^2 \cdot s) \times 1$
273	8.42
283	16.8
293	32.0
303	58.4
308	77.7
313	102
323	174
333	285
343	455
353	708
363	1070
373	1590

Reference

- 4) B.J. Wiersma, "Assessment of the Potential for Hydrogen Generation During Grouting Operations in the R- and P-Reactor Vessels", SRNL-STI-2009-00639, Rev2 May 2010.

Title of Project C Reactor Disassembly Basin H₂

Project No. _____

Subject _____

Works _____

Computer BTWiersmaDate 10/25/11Sheet No. 4

Determination of Critical Areal Density Ratio

$$\left(\frac{A_{Al}}{A_{floor}} \right)_c = \frac{u_0}{Q'_{H_2}}$$

Determine A_{Al}/A_{floor} as a function of temperature. The values for 60% LFL and 100% LFL will be compared for grout and $Ca(OH)_2$. See "C-basin H₂ Generation" spreadsheet, Worksheet VTD calcs.

Areal Density Ratio for Grout

T(°C)	$(A_{Al}/A_{floor})_c$	$(A_{Al}/A_{floor})_c$
	60% LFL	100% LFL
0	2393	5149
10	1199	2580
20	630	1355
30	345	743
35	259	558
40	197	423
50	116	249
60	71	152
70	44	95
80	28	61
90	19	40
100	13	27

Areal Density Ratio for $Ca(OH)_2$

T(°C)	$(A_{Al}/A_{floor})_c$	$(A_{Al}/A_{floor})_c$
	60% LFL	100% LFL
0	508	1094
10	255	548
20	134	288
30	73	158
35	55	119
40	42	90
50	25	53
60	15	32
70	9	20
80	6	13
90	4	9
100	3	6

Determination of Amount of Aluminum
Summary of Reference 2

Area	A_{Al} (ft ²)
Horizontal Tube Storage	9202
Machine Basin	77
Dry Cave	394*
Vertical Tube Storage	3114
D&E Canal	70

* All of Al is in the center pen adjacent to the Dry Cave

Title of Project C- Reactor Disassembly Basin H₂

Project No. _____

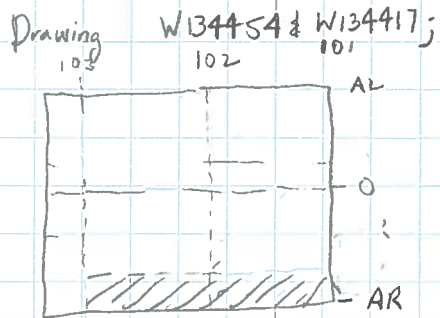
Subject _____

Works _____

Computer B.J. WiersmaDate 10/25/11Sheet No. 5

Cross-Sectional Areas [See Appendix 2 for e-mails from W.B. Griffin]

Vertical Tube Storage (VTS)

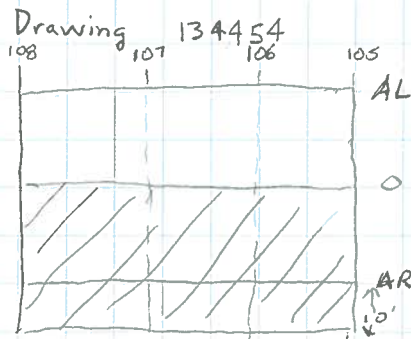


Components were relocated to 'canal' on southside of VTS

- Semi-permanent sleeves are aluminum; Represents ~ 41% of aluminum surface area (182,409 in²)
- Control Rods located in this area; Represents ~ 45% of aluminum surface area (202,718 in² out of 448,425 in²) in VTS
- 2 of 3 USAs located in area 18,912 sq. inches
- Estimated 25 orifice sleeves (~25% of that in all VTS); 8133 in²
- Estimated 448,425 in² in area; ~ 94%
- Assume all aluminum in this area
- Control rods and semi-permanent sleeves are laying horizontal at -30'.

$$\begin{aligned} \text{Area} &= 9' \times 65' \\ &= \underline{585 \text{ ft}^2} \end{aligned}$$

Machine Basin (MB)



10' south of AR to O; 105 to 108

AR to O is 32 ft; 105 to 108 is 62.17 ft

- Safety Rod Thimble and Septifail located in this region

$$\begin{aligned} \text{Area} &= 32 \times 62.17 \\ &= \underline{1989 \text{ ft}^2} \end{aligned}$$

Title of Project C- Reactor Disassembly Basin H₂

Project No. _____

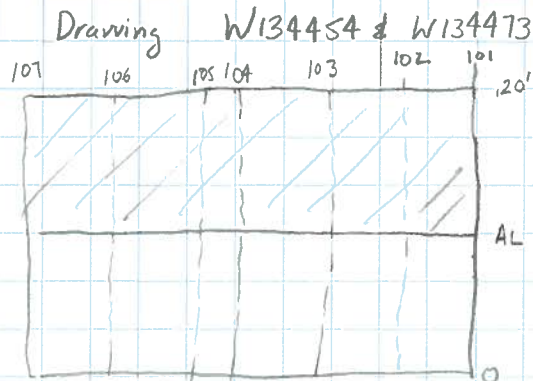
Subject _____

Works _____

Computer B.J. WiersmaDate 10/25/11Sheet No. 6

Cross-Sectional Areas (cont.)

Horizontal Tube Storage (HTS)



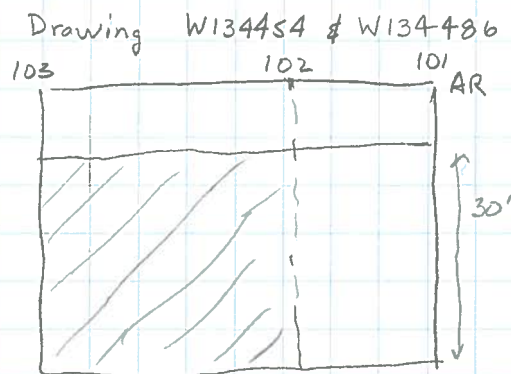
AL to 20' North of AL; 101 to 107

- Slotted Rack in this area;
1,222,144 sg. in represents 92% of Aluminum in HTS

- Assume all aluminum is in this region of the HTS

$$\begin{aligned} \text{Area} &= 20' \times 109' \\ &= \underline{2180 \text{ ft}^2} \end{aligned}$$

Dry Cave (DC)



30 foot (south of AR); 102 to 103

- 6 USH's located in center pen area

$$\begin{aligned} \text{Area} &= 30' \times 24.5' \\ &= \underline{735 \text{ ft}^2} \end{aligned}$$

Title of Project C- Reactor Disassembly Basin H₂

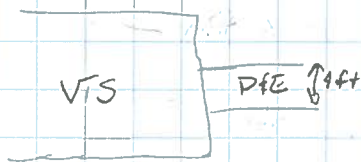
Project No. _____

Subject _____

Works _____

Computer BTWiersmaDate 10/25/11Sheet No. 7

D & E Canal (Drawings W134454, W134417)



64 ft in length

$$\begin{aligned} \text{Area} &= 64 \text{ ft} \times 4 \text{ ft} \\ &= 256 \text{ ft}^2 \end{aligned}$$

Actual Areal Density

<u>Area</u>	<u>A_{AI} / A_{floor}</u>
D & E Canal	0.27
Vertical Tube Storage	5.32
Dry Cave	0.54
Horizontal Tube Storage	4.22
Machine Basin	0.04

Heat Transfer Calculations

Determine the final Temperature of the water and grout after five foot lift has been poured.

Mass of grout and water

ρ_g	Density of grout	125 lb/ft ³	reference 3
ρ_w	Density of water	62.4 lb/ft ³	

Depends on pour height, h

$$m_{\text{grout}} = A_{\text{floor}} h \times \rho_g$$

water For Horizontal Tube Storage (Note: Depth = 17 ft - 6.5833 = 10.4167 ft)

For all others Depth = 30 ft - 6.5833 = 23.4167 ft

$$m_w = A_{\text{floor}} (\text{Depth} - h) \times \rho_w$$

Title of Project C - Reactor Disassembly Basin H₂

Project No. _____

Subject _____

Works _____

Computer BJWiersmaDate 10/25/11Sheet No. 8 C_p - Specific Heat See reference 3

$$C_{p,g} = 0.25 \text{ BTU/(lb} \cdot \text{°F)}$$

$$C_{p,w} = 1.0 \text{ BTU/(lb} \cdot \text{°F)}$$

Temperature

Grout Temperature = T_g = Assume constant @ maximum value of 50°C (120°F)Water Temperature = T_w = Initial ambient temperature of 27°C (80°F)

Analysis Methodology

1st Law of Thermodynamic Energy Balance

$$\begin{array}{ccccccc} \text{Heat} & & \text{Work} & & \text{Internal} & \text{Kinetic} & \text{Potential} \\ & & & & \text{energy} & \text{energy} & \text{energy} \\ Q & - & W & = & \Delta U & + & \Delta KE & + & \Delta PE \end{array}$$

$Q = \Delta U$

$$\Delta U = m C_p (T_f - T_i)$$

 T_f = Final Temperature T_i = Initial Temperature

Q

Assumed same heat transfer as reference 3

$$Q = \bar{Q} \times P_g \times A_{\text{floor}} \times h$$

$$\bar{Q} = 40 \text{ BTU/lb}_m$$

$$Q = \underset{\substack{\downarrow \text{grout}}}{\Delta U_g} + \underset{\substack{\downarrow \text{water}}}{\Delta U_w}$$

Title of Project C - Reactor Disassembly Basin H₂

Project No. _____

Subject _____

Works _____

Computer BTWDate 10/25/11

 Sheet No. 9

$$T_f = \frac{[Q + m_g C_{p,g} T_{i,g} + m_w C_{p,w} T_{i,w}]}{[m_g C_{p,g} + m_w C_{p,w}]}$$

Calculations were performed on spreadsheet "C-basin Temperature" worksheet "Sheet 1"

Results

Pour height, H (ft)	Horizontal Tube Storage Temperature (°C)	All other Areas Temperature °C
0.5	29	28
1.0	32	29
1.5	35	30
2.0	38	32
2.5	42	33
3.0	45	34
3.5	49	36
4.0	53	37
4.5	57	38
5.0	62	40

Maximum Temperatures

Grout - 50°C

Water - Horizontal Tube Storage : 62°C
 All other Areas : 40°C

Title of Project C-Reactor Disassembly Basin H₂

Project No. _____

Subject _____

Works _____

Computer B.J. WiersmaDate 10/25/11Sheet No. 10

Model for Determining Minimum Building Ventilation

- Building can be modeled as a continuous stirred tank reactor to assess the accumulation of H₂ in the vapor space above the basin

$$C_A V_o - C_{Aout} V_o + R_A V_b = \frac{d(C_A V_b)}{dt}$$

C_A = Concentration of H₂ flowing into building = 0

C_{Aout} = Concentration of H₂ flowing out building

R_A = Hydrogen generation rate due to aluminum corrosion

V_o = ventilation rate

V_b = Volume of vapor space above the basin

$$- C_{Aout} V_o + R_A = V_b \frac{dC_A}{dt}$$

Re-write equation in terms of volumetric generation rate and volume fraction of H₂. Use ideal gas law:

$$C_{Aout} = C x_{Aout} \quad \text{where } x_{Aout} = \text{volume fraction of H}_2$$

$$R_A = \frac{dn_A}{dt} = \frac{P}{RT} \frac{dV_A}{dt} \quad \text{where } \frac{dV_A}{dt} = \text{volumetric hydrogen generation rate}$$

$$C = \frac{n}{V} = \frac{P}{RT}$$

$$- \frac{P}{RT} x_{Aout} V_o + \frac{P}{RT} \frac{dV_A}{dt} = V_b \frac{P}{RT} \frac{dx_A}{dt}$$

$$\frac{dx_A}{dt} + \frac{V_o}{V_b} x_A = \frac{dV_A}{dt} \frac{1}{V_b}$$

Title of Project C-Reactor Disassembly Basin H₂

Project No. _____

Subject _____

Works _____

Computer B.J. WiersmaDate 10/25/11Sheet No. 11

SRS

- $\frac{dV_A}{dt}$ changes with time due to passivation of aluminum

- Assume $\frac{dV_A}{dt}$ is constant. Assume the rate is equal to the hydrogen generated in one day divided by the hours in a day.

- From reference 4.

$$\frac{dV_A}{dt} = \frac{\int_0^t Q'_{H_2} e^{-2.31t} dt}{\Delta t}$$

Q'_{H_2} = Volumetric H₂ generation rate per unit Area
t = time in hours

$$\begin{aligned} \frac{dV_A}{dt} &= - \frac{Q'_{H_2}}{2.31} \left[e^{-2.31t} \right] \Big|_0^{24} \\ &= \frac{Q'_{H_2}}{2.31(24)} \left[1 - e^{-2.31(24)} \right] \\ &= - \frac{Q'_{H_2}}{55.4} \end{aligned}$$

In order to obtain a volumetric flow rate Q'_{H_2} must be multiplied by the surface area of aluminum reacting. As a conservative estimate it will be assumed that all the aluminum in the basin is reacting. Reference 2 indicates that this is $A_A = 12,838 \text{ ft}^2$.

$$R'_A = \frac{dV_A}{dt} = \frac{Q'_{H_2}}{55.4} \times A_{A1} = 209 \times Q'_{H_2}$$

Title of Project

C-Reactor Disassembly Basin H₂

Project No.

Subject

Works

Computer B.J. Wiersma

Date

10/25/11

Sheet No. 12

SRS

For $\text{Ca}(\text{OH})_2$ the highest Q'_{H_2} could occur in the horizontal tube storage area where the temperature could rise to 62°C . $Q'_{\text{H}_2} = 2.85 \times 10^{-5} \frac{\text{m}^3}{\text{m}^2\text{-s}}$ from the table on sheet 3.

For grout at a temperature of 50°C , $Q'_{\text{H}_2} = 3.69 \times 10^{-6} \frac{\text{m}^3}{\text{m}^2\text{-s}}$

Solve initial value problem

$$\frac{dx_A}{dt} + \frac{V_0}{V_b} x_A = \frac{R'_A}{V_b}$$

$$x_A = 0 \quad \text{at} \quad t = 0$$

$$x_A = \frac{R'_A}{V_0} \left(1 - e^{(-tV_0/V_b)} \right)$$

$V_b =$ Volume of vapor space above basin

Cross-sectional Area per drawing W134454

$$203.25 \text{ feet} \times 130 \text{ feet} = 26,423 \text{ ft}^2$$

Height ≈ 20 feet above the water level of the basin

$$V_b = 528,450 \text{ ft}^3$$

- Determine x_A as a function of v_0 .
- Goal is to keep x_A less than 60% of the LFL.

APPENDIX 2



Re: 105-C D&E Canal in Process Room 

William Griffin to: Bruce Wiersma

Cc: John Blankenship, John Musall, Michael Serrato, Christine
Langton, Kristine Zeigler, Brenda Garcia-Diaz

06/22/2011 04:34 PM

History: This message has been replied to.

See comments in red below; slight variation to yours.

Bill Griffin
D&D Engineer
Office 803-952-6449
Cell 803-761-1338
Pager 31164

Bruce Wiersma

Bill, I thought I would write an e-mail to verify our...

06/22/2011 12:18:59 PM

From: Bruce Wiersma/SRNL/Srs
To: William Griffin/SRNS/Srs@Srs
Cc: John Blankenship/SRNS/Srs@Srs, John Musall/SRNS/Srs@Srs, Michael Serrato/SRNL/Srs@Srs,
Christine Langton/SRNL/Srs@Srs, Kristine Zeigler/SRNL/Srs@Srs, Brenda
Garcia-Diaz/SRNL/Srs@Srs
Date: 06/22/2011 12:18 PM
Subject: Re: 105-C D&E Canal in Process Room

Bill,

I thought I would write an e-mail to verify our discussions from yesterday. We were trying to determine a representative local area for the analysis. That is what is the cross-sectional area of the basin that should be considered for the analysis. I have summarized what we said by referring to coordinates that appear on drawing W134454.

Vertical Tube Storage: AR to O by 101 to 102; Control Rods contribute to majority of aluminum surface area. **Agree**

Machine Basin: AR to O and 106 to 108; Septifoil and Safety Rod Thimble contribute to majority of aluminum surface area. **10' south of AR to O by 105 to 108**

Horizontal Tube Storage: AL to 18 feet north of AL by 101 to 107; Racks with USH's contributes to the majority of the surface area. **AL to 20' north of AL by 101 to 107**

Dry Caves: 28.5 feet by 102 to 103; USH's present. **30' by 102 to 103**

D&E Canal: 64 feet by 4 feet; We had agreed on this previously. **Agree**


If you could let me know if you agree with this.

Thanks,

Bruce J. Wiersma
Savannah River National Laboratory
Materials Science and Technology
Bldg. 773-A, Rm. D-1125
Phone: 725-5439
FAX: 725-7369
e-mail: bruce.wiersma@srnl.doe.gov

William Griffin	I just sent you a copy. Bill Griffin	05/23/2011 04:29:11 PM
Bruce Wiersma	Bill, Thanks for the photos. I was looking for the...	05/23/2011 04:23:46 PM
William Griffin	Photos for your use and info. Bill Griffin	05/23/2011 04:18:56 PM



Re: C-Basin Hydrogen Calculation Revision 
William Griffin to: Bruce Wiersma
Cc: John Blankenship, John Musall

10/18/2011 01:55 PM

History: This message has been replied to.

The majority of aluminum components (90% +) that were in the referenced area were relocated to the long canal on the south side of VTS and are now laying on the bottom of that area; no aluminum components are stored vertically in the basin. This canal is 9' wide by 65' long or 585 sq. ft. Let me know if you need any additional info.

Bill Griffin
D&D Engineer
Office 803-952-6449
Cell 803-761-1338
Pager 31164

Bruce Wiersma

Bill, I am in the process of revising the document...

10/18/2011 12:23:30 PM

From: Bruce Wiersma/SRNL/Srs
To: William Griffin/SRNS/Srs@Srs
Date: 10/18/2011 12:23 PM
Subject: C-Basin Hydrogen Calculation Revision

Bill,

I am in the process of revising the document. I wanted to confirm something with you. The previous cross-sectional area that we used for VTS was 664 sq. ft. Based on drawing w134454 the area was bounded by AR to O; and 101 to 102. Is this still valid or is the area more restricted?

If I have this information by tomorrow, I think I should be able to have a draft for your review by Monday.

Thanks,

Bruce J. Wiersma
Savannah River National Laboratory
Materials Science and Technology
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FAX: 725-7369
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