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**ASSESSMENT OF THE POTENTIAL FOR HYDROGEN GENERATION
DURING DEACTIVATION AND DECOMMISSIONING OF REACTOR
VESSELS AT THE SAVANNAH RIVER SITE - 11197**

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ABSTRACT

The R- and P-reactor vessels at the Savannah River Site (SRS) are being prepared for deactivation and decommissioning (D&D). D&D activities will consist primarily of physically isolating and stabilizing the reactor vessel by filling it with a grout material. The reactor vessels contain aluminum alloy materials, which pose a concern in that aluminum corrodes rapidly when it comes in contact with the alkaline grout. A product of the corrosion reaction is hydrogen gas and therefore potential flammability issues were assessed.

A model was developed to calculate the hydrogen generation rate as the reactor is being filled with the grout material. Three options existed for the type of grout material for D&D of the reactor vessels. The grout formulation options included ceramcrete (pH 6-8), a calcium aluminate sulfate (CAS) based cement (pH 10), or Portland cement grout (pH 12.4). Corrosion data for aluminum in concrete were utilized as input for the model. The calculations considered such factors as the surface area of the aluminum components, the open cross-sectional area of the reactor vessel, the rate at which the grout is added to the reactor vessel, and temperature. Given the hydrogen generation rate, the hydrogen concentration in the vapor space of the reactor vessel above the grout was calculated. This concentration was compared to the lower flammability limit for hydrogen.

The assessment concluded that either ceramcrete or the CAS grout may be used to safely grout the P-reactor vessel. The risk of accumulation of a flammable mixture of hydrogen between the grout-air interface and the top of the reactor is very low. Portland cement grout, on the other hand, for the same range of process parameters did not provide a margin of safety against the accumulation of flammable gas in the reactor vessel during grouting operations in the P-reactor vessel. Therefore, it was recommended that this grout not be utilized for this task.

On the other hand, the R-reactor vessel contained significantly less aluminum surface area than the P-reactor vessel based on current facility process knowledge, surface observations, and drawings. Therefore, a Portland cement grout may be considered for grouting operations as well as the other grout formulations.

INTRODUCTION

Reactor facilities were constructed at the Savannah River Site (SRS) to produce materials for the nation's nuclear weapons stockpile. The central component of the facility was the reactor vessel where the materials were produced. The reactor vessel was classified as a heavy water production reactor, in which the moderator water is used both for cooling and moderating the nuclear reaction. The vessel was constructed of stainless steel. However, the internal components, such as the universal sleeve housings, septifoils, spargers, and thimbles, were all constructed of aluminum alloys. Figure 1 shows the configuration of these internal components.

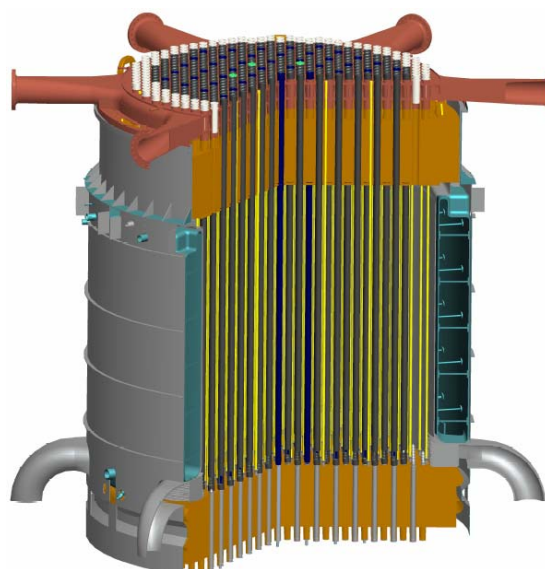


Figure 1. Cross-sectional view of a reactor vessel.

The R- and P-reactors at the SRS were removed from service in 1964 and 1988, respectively, and are now being prepared for deactivation and decommissioning (D&D). The closure technique for the reactor buildings is called In-Situ Decommissioning (ISD) [1]. The technique consists of placing cementitious grout materials below grade up to the ground surface. The above grade structure will then be demolished and removed. Finally, a concrete cap will cover the area and this will be the final configuration.

The internal aluminum components pose a concern in that aluminum corrodes very rapidly when it comes in contact with the very alkaline grout materials, and as a result produces hydrogen gas. To address this potential deflagration/explosion hazard, existing experimental and analytical studies of this issue were evaluated to determine if any process constraints on the chemistry of the fill material and the fill operation are necessary.

Various options exist for the type of grout material that may be used for D&D of the reactor vessels. The grout formulation options include ceramicrete (pH 6-8), a specially

designed calcium aluminate sulfate (CAS) based cement (pH 10) [2], or portland cement grout (pH 12.5). To perform this task, the rate of hydrogen generation in the vessels for grouts with a pH range from 8 to 13 were calculated. The calculations considered such factors as temperature, the rate at which the grout fills the vessel, the surface area of the components present, the surface area of the reactor vessel and the void volume of the reactor vessel. The objective of these calculations was to provide input as to which grout formulation is appropriate for the operations so that the risk of hydrogen gas accumulation is minimized.

ANALYTICAL APPROACH

A similar evaluation was performed for the K basins at the Hanford Site [3]. This analysis was reviewed and applied to the situation for the R- and P- reactor vessels. The process is as follows:

- 1) Aluminum corrodes upon exposure to the grout.
- 2) Hydrogen is generated as a consequence of the corrosion reaction.
- 3) The gas rises to the surface of the grout in the form of bubbles.
- 4) The bubbles will burst at the grout 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, pH, fill rate 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 [3].

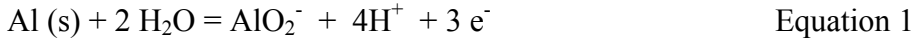
The assumptions used in the analysis were:

- The aluminum is exposed to wet cement while the reactor vessel is being filled. Corrosion and hydrogen generation rates associated with this condition were assumed.
- There are openings in the reactor vessel that allow hydrogen to escape the vessel.
- Once the hydrogen reaches the top of the reactor vessel, there is sufficient advection to disperse the hydrogen within the building superstructure.

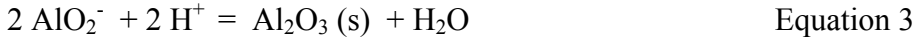
Based on these assumptions the only place that hydrogen could potentially accumulate is in the region between the grout layer and the top of the reactor vessel.

The first part of the model involved developing a kinetic expression for the generation of hydrogen due to aluminum corrosion. The corrosion mechanism for aluminum in an

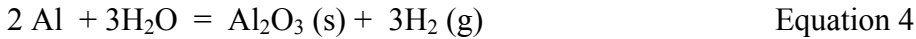
alkaline environment is represented the anodic and cathodic reactions shown in Equations (1) and (2), respectively.



At low concentrations and pH between 8 and 14, the aluminate ion (AlO_2^-) is unstable and will tend to form an aluminum oxide phase according to Equation 3.



Therefore, the net reaction for corrosion of aluminum is:



Equation 4 shows that for every mole of aluminum that corrodes, 1.5 moles of hydrogen evolves.

This kinetic expression can be represented by the following relationship:

$$Q_o = f(\text{pH}, T) = U(\text{pH}) * V(T) \quad \text{Equation 5}$$

Where Q_o is the hydrogen generation rate in $\text{cm}^3/\text{cm}^2/\text{min}$, T is the grout temperature in K, U is the hydrogen generation rate as a function of pH and V is the hydrogen generation rate as a function of temperature.

Literature values for the corrosion rate of aluminum as a function of pH were obtained for Portland cements, CAS cements, cement pore water solutions, and sodium hydroxide solutions [3-6]. These values were reported as either corrosion rates or hydrogen generation rates. Figure 2 shows the hydrogen generation rates from these literature values as a function of pH. These rates were measured shortly after the aluminum was exposed and at approximately 30 °C. The figure shows that the hydrogen generation rate increases exponentially as the pH increases according to Equation 6.

$$U = 3 \times 10^{-14} \exp(2.1 * \text{pH}) \quad \text{Equation 6}$$

where U is the hydrogen generation rate as a function of pH in $\text{cm}^3/\text{cm}^2/\text{min}$.

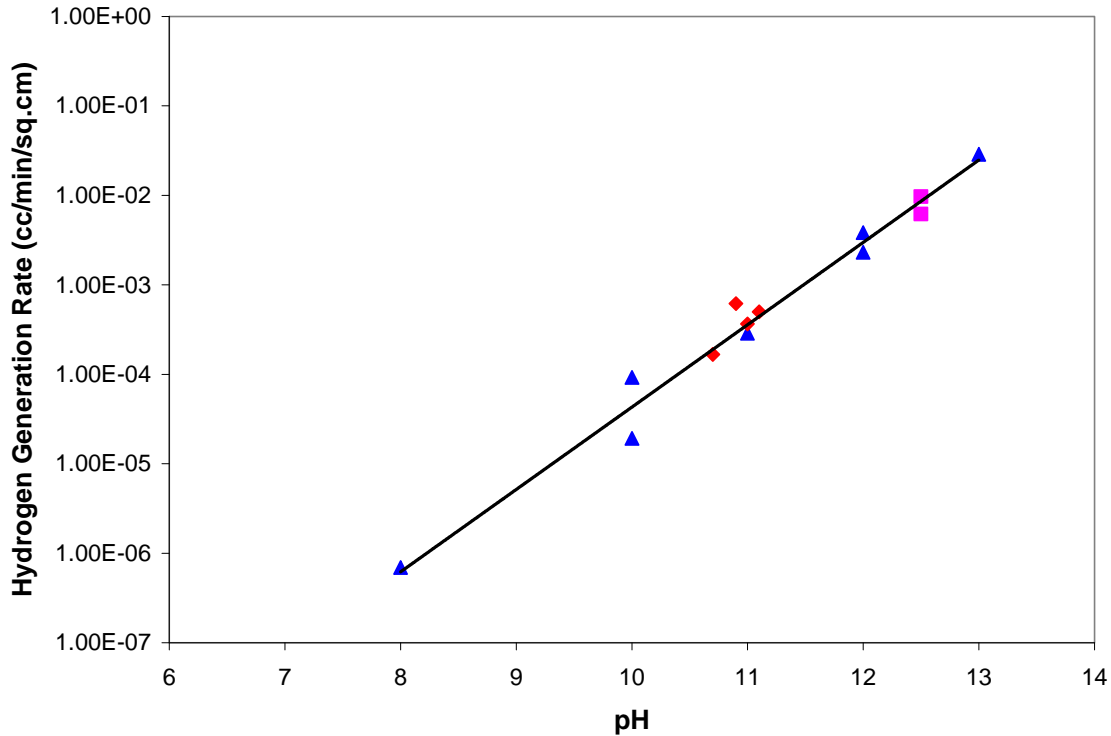


Figure 2. Hydrogen generation rate as a function of the pH. Blue triangles are literature data for the initial rates with aluminum exposed to calcium hydroxide and sodium hydroxide solutions [4, 5]. Red diamonds are literature data for the initial rates with aluminum exposed to various CAS grout materials [6]. Purple squares are literature data for the initial rates with aluminum exposed to Portland cement grout materials [3, 6].

The temperature dependence of the hydrogen generation rate was also determined from experimental data in the literature. Two assumptions were made in order to develop a relationship between the hydrogen generation and temperature. First, it was assumed that the hydrogen generation could be described by the Arrhenius expression. Secondly, hydrogen generation rates measured in calcium hydroxide solutions that simulate the grout pore water are appropriate estimates for actual grout. The hydrogen generation rate in calcium hydroxide solutions increased by a factor of 5 as the temperature increased from 23 °C to 52 °C [3]. This result was consistent with the temperature dependence that was observed in an inhibited grout mixture [7]. Utilizing this data in the Arrhenius expression, the activation temperature was determined to be 5339 K. Thus, the final relationship for the temperature dependence may be expressed as:

$$V = A \exp (-5339/T) \quad \text{Equation 7}$$

where V is the hydrogen generation rate as a function of temperature in $\text{cm}^3/\text{cm}^2/\text{min}$, A is a constant, and T is the temperature in K.

Substitution of Equations 6 and 7 into Equation 5 yields:

$$Q_0 = 3 \times 10^{-14} \exp(2.1 \cdot \text{pH}) \cdot \exp(-5339 \cdot (1/T - 1/303)) \quad \text{Equation 8.}$$

The total hydrogen generated (Q) also depends on the surface area of aluminum that is actively corroding. Laboratory tests indicated that the initial hydrogen generation rate decreases by 50% every three hours [3]. 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 half-life for the hydrogen generation rate, $Q_{1/2}$, may be expressed as:

$$Q_{1/2} = Q_0 \cdot \exp(-0.231 \cdot t) \quad \text{Equation 9}$$

where t is the time in hours. A material balance was performed at time t_f to determine the hydrogen generated from the surface area of aluminum that is exposed to the grout mixture. Equation 10 represents this material balance.

$$Q A_c H = \int_0^H Q_0 (\exp(-0.231 t_f)) (A_c (dh)) \quad \text{Equation 10}$$

where Q is the total hydrogen gas released due to corrosion, A_c is the open cross-sectional area of the vessel, H is the height of the reactor vessel, h is the height above the bottom of the reactor vessel. For the calculations it was assumed that the grout fill level in the reactor increases as a linear function of time. Therefore, t_f is related to the fill height by the following:

$$t_f = h/m \quad \text{Equation 11}$$

where m is the grout fill rate in cm/min.

The solution to Equation 10 is therefore,

$$Q = \frac{Q_0 m}{0.00385 H} \left(1 - \exp\left(-0.00385 \frac{H}{m}\right) \right) \quad \text{Equation 12}$$

The second part of the model involves the mass transport of the hydrogen gas from the surface of the grout to the top of the reactor. The transport of the relatively light hydrogen gas through the dense air layer above the grout is a process analogous to Fickian diffusion. The concept of a vertical turbulent diffusion has been applied in modeling the upward transport of a lighter fluid through a heavier miscible fluid. Epstein and Burelebach developed the diffusion equation and boundary conditions for a brine/water turbulent diffusion layer [8]. By analogy these equations may be applied to mixing in a heavy gas/ light gas system. The analysis does not account for dissipation of hydrogen between the surface of the grout and the top of the reactor due to advection. Accounting for this phenomenon would minimize the accumulation of hydrogen in this region.

Solution of the diffusion equation allows one to calculate the flux of the hydrogen away from the grout surface. This flux is related to the superficial velocity, u_o , which may be expressed as:

$$u_o = \beta^2 * [g * H_o * (1 - (M_{H_2}/M_{air})) * X_{LFL}^3]^{1/2} \quad \text{Equation 13}$$

where β is a proportionality constant, g is the acceleration of gravity, H_o is the distance between the grout-air interface and the top of the reactor, M is the molecular weight of either hydrogen or air, and X is the volume % of hydrogen in air at the lower flammability limit (LFL). In the case of hydrogen the LFL is 4% by volume.

The incipient flammability condition occurs when the gas generation rate due to corrosion equals the flux of hydrogen through air. The boundary condition at the interface between the grout and air is that the hydrogen gas concentration is at the LFL. For safety class operations, with radioactive materials stored within a vessel, a criterion of 60% LFL was utilized for the evaluation. The equation that describes this condition is:

$$Q * A_{Al} = u_o * A_c \quad \text{Equation 14}$$

where A_{Al} is the surface area of aluminum in contact with grout and A_c is the open cross-sectional area of the reactor vessel. Equation 14 can be re-arranged to give the critical areal density ratio.

$$[A_{Al}/A_c]_c = u_o/Q \quad \text{Equation 15}$$

For the analysis, a plot of $[A_{Al}/A_c]_c$ vs. H is prepared (see Figure 3). If the critical area density is greater than the actual areal density, there is a low probability of a flammable condition. On the other hand if it is less than the actual areal density, there is a possibility of a flammable condition developing.

DETERMINATION OF AREAL DENSITY RATIOS

Actual areal density ratios are being calculated based on current facility process knowledge, surface observations, and drawings of the R- and P-reactor vessels. The predominant aluminum components present in the reactor vessels are the universal sleeve housing (USH) and thimble tubes. It will be assumed that the inner and outer surfaces of these components will be exposed to the grout. The aluminum surface area, A_{Al} , as a function of the fill level, H , was calculated from the following relationship:

$$A_{Al}(h) = N_{USH} * \pi * (D_{USH_o} + D_{USH_i}) * H + N_T * \pi * (D_{T_o} + D_{T_i}) * H \quad (7)$$

where D is the diameter, subscript USH is for the USH tubes, subscript T is for the thimble tubes, subscript o represents the exterior surface, subscript i represents the

interior surface, and N is the quantity of USH or thimble tubes. The calculation did not include the surface area of the ends of the tubes.

The open cross-sectional area of the vessel, A_c , was calculated by subtracting the cross-sectional area of the USHs, thimble tubes, septifoils and spargers from the total tank cross-sectional area. This is represented by the following equation:

$$A_c = \pi * D_t^2/4 - N_{USH} * \pi * (D_{USH_o}^2 - D_{USH_i}^2)/4 - N_T * \pi * (D_{T_o}^2 - D_{T_i}^2)/4 - N_{ss} * \pi * D_{ss}^2/4 \quad (8)$$

where subscript c is for the cross-sectional area, subscript t is for the tank, and subscript ss is for the septifoils and the spargers. In P-reactor it is estimated that there are 432 USH tubes, 61 septifoils, and 66 thimble tubes and 6 spargers. The USH tubes have outer and inner diameters, 10.8 centimeters and 10.1 centimeters, respectively. The outer and inner diameters for the thimble tubes are 3.8 centimeters and 2.5 centimeters, respectively. The septifoils and spargers were modeled as a cylinder with a diameter of 8.9 centimeters. In R-reactor, it was determined that there are no USH tubes present (i.e., only septifoils, thimbles and spargers are present). The lack of USH tubes means that A_{Al} , and hence the areal density ratio, for R-reactor is less than that of P-reactor by a factor of approximately 25 (see Figure 3).

RESULTS

Case studies were performed to evaluate the effects of the grout temperature, fill rate, and grout pH on the hydrogen generation rate. The cases are summarized in Table 1 and an example of a critical. Some of the key trends were:

- An increase in temperature resulted in a lower critical areal density and therefore greater risk of developing a flammable condition.
- An increase in pH resulted in a lower critical areal density and therefore a greater risk of developing a flammable condition (see Figure 3).
- An increase in the fill rate resulted in a lower critical areal density, although the effect was not as great as temperature or pH.
- The 60% LFL criterion provides a significant margin on the risk of developing a flammable condition.
- An increase in the actual areal density ratio results in a greater risk of developing a flammable condition. Thus, as shown in the figures, there is a greater risk of developing a flammable condition in the P-reactor vessel than there is in the R-reactor vessel.

The results of the case studies for the P-reactor vessel demonstrate that two of the grout formulations, the ceramicrete and the CAS, should not result in a flammable condition during reactor vessel grouting operations as long as they are within the parameters of the case studies. Figure 3 shows the base case that was considered (i.e., 70 °C, fill rate of 2.5 cm/min, 60% LFL). Even at 60% LFL the critical areal density ratio for the CAS grout is at least 1 to 2 orders of magnitude greater than the actual areal density ratio, while the ratio for the ceramicrete is 3 to 4 orders of magnitude greater. At 100% LFL these

margins increase further to 2 to 3 orders of magnitude for the CAS grout and 4 to 5 orders of magnitude for the ceramicrete. The portland cement grout is not a viable option for the P-reactor vessel as it exceeds the 60% LFL criterion for each case that was examined. Figure 3 shows that after 120 inches of the pH 12.5 grout has been added, there is a risk that a flammable mixture of gas could form near the surface of the grout.

The results of the case studies for the R-reactor vessel suggest that all three grout formulations may be viable. For example, at the base case conditions (i.e., 70 °C, fill rate of 1 inch/min, 60% LFL, see Figure 3) the critical areal density ratio is a factor of 5.5 greater than the actual areal density ratio for the Portland cement, while the ratio for the CAS grout is 2 to 3 orders of magnitude greater and 4 to 5 orders of magnitude greater for the ceramicrete grout. Although not shown, similar margins would be obtained at a temperature of 80 °C with a fill rate of 1.25 cm/min.

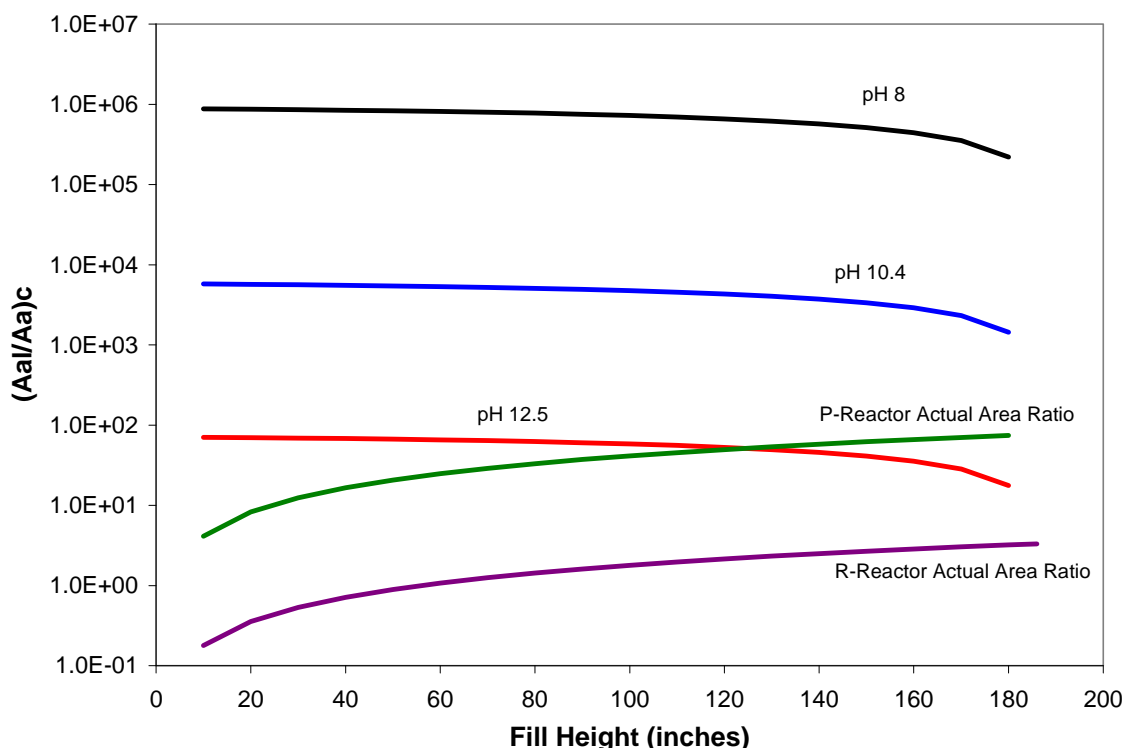


Figure 3. Flammability evaluation for the P- and R-reactor vessels exposed to grouts with different pH.

The factor of 5.5 difference between the critical areal density ratio and the actual areal density was examined further to assess the risk associated with utilizing Portland cement grout for R-reactor. The surface area of the aluminum thimbles was approximately 620,800 square centimeters. A total aluminum surface area of approximately 3,415,000 square centimeters could be contained in the R-reactor vessel before the 60% LFL criterion would be exceeded (i.e., there would need to be an additional 2,800,000 square centimeters of aluminum in the reactor vessel). This surface area is roughly equivalent

to 90 USH's. Thus, if the facility is confident in the information provided by current facility process knowledge and drawings, the risk of approaching the LFL is low.

Although these results are encouraging, due to the potential consequences, taking precautions that reduce the likelihood of a flammable condition are recommended. These measures include ensuring that the building has adequate ventilation during the grouting process, minimizing the grout temperature, and operating at a slower fill rate. In order to evaluate the ventilation needs, the volumetric flow rate of hydrogen was calculated for each case for the P and R-reactor vessels utilizing Equation 12. The results are summarized in Tables 2 and 3.

For the P-reactor vessel, the hydrogen flow rates for the pH 8 and pH 10.4 grout are very small, less than 28 liters/min (see Table 2). The flow rates for the pH 12.5 grout are higher than those for the lower pH grouts, ranging between 156 and 2230 liters/min, however, the rates could be manageable with proper ventilation. For the R-reactor vessel (see Table 3), the flow rates are a factor of approximately 25 less than the P-reactor vessel (i.e., proportional to the change in aluminum surface area).

Table 1. Summary of Case Studies

Case	Temperature (°C)	Fill Rate (cm/minute)	pH
1	50	2.5	8, 10.4, and 12.5
2	100	2.5	8, 10.4, and 12.5
3	70	2.5	8, 10.4, and 12.5
4	80	2.5	8, 10.4, and 12.5
5	50	1.25	8, 10.4, and 12.5
6	60	1.25	8, 10.4, and 12.5
7	70	1.25	8, 10.4, and 12.5
8	50	5	8, 10.4, and 12.5
9	60	5	8, 10.4, and 12.5
10	70	5	8, 10.4, and 12.5
11	100	5	8, 10.4, and 12.5

Table 2. Summary of Volumetric Flow Rates of Hydrogen for the P- Reactor Vessel Case Studies.

Case	Temperature (°C)	Fill Rate (cm/min)	pH 10.4	pH 8	pH 12.5
			Qtot (liters/min)	Qtot (liters/min)	Qtot (liters/min)
1	50	2.5	2.55E+00	1.67E-02	2.12E+02
2	100	2.5	2.33E+01	1.53E-01	1.90E+03
3	70	2.5	6.68E+00	4.25E-02	5.43E+02
4	80	2.5	1.04E+01	6.79E-02	8.43E+02
5	50	1.25	1.92E+00	1.25E-02	1.56E+02
6	60	1.25	3.14E+00	2.07E-02	2.56E+02
7	70	1.25	4.98E+00	3.28E-02	4.08E+02
8	50	5	2.97E+00	1.95E-02	2.43E+02
9	60	5	4.90E+00	3.11E-01	3.99E+02
10	70	5	7.81E+00	5.09E-02	6.37E+02
11	100	5	2.73E+01	1.78E-01	2.23E+03

Table 3. Summary of Volumetric Flow Rates of Hydrogen for the R- Reactor Vessel Case Studies.

Case	Temperature (°C)	Fill Rate (cm/min)	pH 10.4	pH 8	pH 12.5
			Qtot (liters/min)	Qtot (liters/min)	Qtot (liters/min)
1	50	2.5	1.13E-01	7.39E-04	9.17E+00
2	100	2.5	1.02E+00	6.79E-03	8.41E+01
3	70	2.5	2.83E-01	1.93E-03	2.41E+01
4	80	2.5	4.58E-01	3.00E-03	3.74E+01
5	50	1.25	8.46E-02	5.52E-04	6.88E+00
6	60	1.25	1.39E-01	9.08E-04	1.13E+01
7	70	1.25	2.22E-01	1.45E-03	1.80E+01
8	50	5	1.32E-01	8.63E-04	1.08E+01
9	60	5	2.18E-01	1.42E-03	1.77E+01
10	70	5	3.45E-01	2.26E-03	2.82E+01
11	100	5	1.22E+00	7.92E-03	9.85E+01

CONCLUSIONS

An assessment of the potential for hydrogen generation during grouting operations in the R- and P- Reactor vessels was performed. The assessment concluded that either ceramicrete or the CAS grout may be used to safely grout the P-reactor vessel because neither grout will generate enough hydrogen to exceed 60% LFL. The risk of accumulation of a flammable mixture of hydrogen between the grout-air interface and the top of the reactor is very low. Portland cement grout, on the other hand, for the same range of process parameters does not provide a significant margin of safety against the accumulation of flammable gas in the reactor vessel during grouting operations in the P-

reactor vessel. This accumulation of flammable gas exceeds 60% LFL. It is recommended that this grout not be utilized for this task.

Based on current facility process knowledge, drawings and surface observations, the R-reactor vessel contains significantly less aluminum than P-reactor. Thus a Portland cement grout may be considered as well. For example, if the grout fill rate is less than 1 inch/min and the grout temperature is maintained at 70 °C or less, the risk of hydrogen accumulation in the R-reactor vessel is low for the Portland cement. Alternatively, if the grout fill rate is less than 0.5 inch/min and the grout is maintained at a temperature of 80 °C, the risk is again low. In either case, accumulation of flammable gas does not exceed 60% LFL.

Although these calculations are conservative, there are some measures that may be taken to further minimize the potential for hydrogen evolution.

1. Minimize the temperature of the grout as much as practical. Lower temperatures will mean lower hydrogen generation rates. For P-reactor, grout temperatures less than 100 °C should provide an adequate safety margin for the pH 8 and pH 10.4 grout formulations. For R-reactor, grout temperatures less than 70 °C or 80 °C will provide an adequate safety margin for the Portland cement. The other grout formulations are also viable options for R-reactor.
2. Minimize the grout fill rate as much as practical. Lowering the fill rate takes advantage of passivation of the aluminum components and hence lower hydrogen generation rates. For P-reactor, fill rates that are less than 2 inches/min for the ceramicrete and the CAS grouts will reduce the chance of significant hydrogen accumulation. For R-reactor, fill rates less than 1 inch/min will again minimize the risk of hydrogen accumulation.
3. Ventilate the building as much as practical (e.g., leave doors open) to further disperse hydrogen. The volumetric hydrogen generation rates in the P-reactor vessel, however, are low for the pH 8 and pH 10.4 grout i.e., less than 28 liters/min.

REFERENCES

- 1) C. A. Langton, M. G. Serrato, J. K. Blankenship, and W. B. Griffin, "Savannah River Site R-Reactor Disassembly Basin In-Situ Decommissioning", WM 2010, Paper No. 10449, Phoenix, AZ, March 7-11, 2010.
- 2) C. A. Langton, et.al., "Use of Cementitious Materials for SRS Reactor Facility In-Situ Decommissioning", WM 2011, Paper No. 11620, Phoenix, AZ, February 27-March 3, 2011.
- 3) S. M. Short and B. M. Parker, "Potential for Generation of Flammable Mixtures of Hydrogen from Aluminum-Grout Interaction During Basin Grouting", PNNL-15156, April 2005.
- 4) Chatalov in Atlas of Electrochemical Equilibria in Aqueous Solutions, M. Pourbaix, NACE, Houston, TX, 1974.

- 5) A. B. McKee and R. H. Brown, "Resistance of Aluminum to Corrosion in Solutions Containing Various Anions and Cations", *Corrosion*, Vol. 3, December, 1947, p. 595.
- 6) M. Hayes and I. H. Godfrey, "Development of the Use of Alternative Cements for the Treatment of Intermediate Level Waste", WM'07 Conference, Tuscon, AZ February 25-March 1 2007.
- 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) 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.

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