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Document Control Desk
U.S. Nuclear Regulatory Commission
Washington, DC 20555-0001

Attention: Mr. Jeffrey A. Ciocco,

Docket No. 52-021
MHI Ref: UAP-HF-09199

Subject: MHI's Second Responses to US-APWR DCD RAI No.234

- References:** 1) "Request for Additional Information No. 234 Revision 1, SRP Section: 06.05.02 - Containment Spray as a Fission Product Cleanup System, Application Section: 6.5.2 Containment Spray Systems," dated February, 26, 2009.
- 2) Letter MHI Ref: UAP-HF-09121 from Y. Ogata (MHI) to U.S. NRC, "MHI's Responses to US-APWR DCD RAI No. 234" dated March 24, 2009

With this letter, Mitsubishi Heavy Industries, Ltd. ("MHI") transmits to the U.S. Nuclear Regulatory Commission ("NRC") a document entitled "Response to Request for Additional Information No. 234-2040 Revision 1".

Enclosed are the second responses to the RAIs contained within Reference 1. In the initial responses submitted with Reference 2, MHI committed to submit responses to RAI #6.5.2-1 and #6.5.2-3 by 26th of April 2009.

Please contact Dr. C. Keith Paulson, Senior Technical Manager, Mitsubishi Nuclear Energy Systems, Inc. if the NRC has questions concerning any aspect of the submittals. His contact information is below.

Sincerely,

Y. Ogata

Yoshiki Ogata,
General Manager- APWR Promoting Department
Mitsubishi Heavy Industries, LTD.

Enclosure:

1. Response to Request for Additional Information No. 234-2040 Revision 1

CC: J. A. Ciocco
C. K. Paulson

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NRC

Contact Information

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Docket No. 52-021
MHI Ref: UAP-HF-09199

Enclosure 1

UAP-HF-09199
Docket No. 52-021

Response to Request for Additional Information No. 234-2040
Revision 1

April 2009

RESPONSE TO REQUEST FOR ADDITIONAL INFORMATION

4/22/2009

US-APWR Design Certification

Mitsubishi Heavy Industries

Docket No. 52-021

RAI NO.: NO. 234-2040 REVISION 1
SRP SECTION: 06.05.02 – CONTAINMENT SPRAY AS A FISSION PRODUCT CLEANUP SYSTEM
APPLICATION SECTION: 6.5.2
DATE OF RAI ISSUE: 2/26/2009

QUESTION NO.: 06.05.02-1

- a. Under DBA conditions, how long does it take for the RWSP water to reach a pH of 7 due to dissolution of NaTB?
- b. What is the dissolution rate of NaTB?

Background

GDC 41 establishes the design requirements for containment atmosphere cleanup systems which function to reduce the concentration and quality of fission products released to the environment following postulated accidents. Compliance with GDC 41 ensures that the containment spray system will accomplish the fission product removal function assumed in the Chapter 15 radiological consequence calculations. To meet this design criterion, fission products must be retained as well as removed by the spray solution.

To control revolution of iodine from the containment sump water following a DBA, SRP 6.5.2 Acceptance Criterion II.1.G (p. 6.5.2-5), recommends that long term iodine retention can only be assured when a pH above 7 is achieved and that this pH should be achieved by the onset of the spray recirculation mode.”

The DCD states that sodium tetra-borate decahydrate (NaTB) is added to the spray/sump solution to raise the pH for iodine retention. The mechanism of addition is that baskets of NaTB are placed in the containment where spray water will fall on them, dissolve the NaTB, and drain back into the sump (RWSP). As configured, only a few of the sprays near the outer periphery of the containment will fall into the NaTB baskets (Figs. 6.3-10 and 6.3-11). The DCD (6.3.2.2.5) states that dissolution time for all NaTB is 12 hours, although no calculation is given. This seems to contradict the SRP 6.5.2 Acceptance Criterion II.1.G (p. 6.5.2-5), which says that a pH of 7 should be achieved by the onset of the spray recirculation mode.

ANSWER:

- a. The NaTB contained in baskets is dissolved in 12 hours. Therefore, if it is expected that re-circulating water is buffered by base of NaTB solution only, it takes longer than 12 hours after

the occurrence of an accident for re-circulating water to reach a pH of 7. However, cesium is released from damaged fuel, and released cesium (CsOH in re-circulating water) raises a pH for re-circulating water. As a result, a pH for re-circulating water reaches to 7 within about 1 hour.

The recirculation water pH was calculated considering factors described below that contribute to the recirculation water pH after an accident.

- (1) Contribution by (initial) contained boric acid immediately after LOCA
- (2) Contribution due to generation of hydrogen iodide (HI) by iodine release in FP (fission product)
- (3) Contribution due to generation of nitric acid (HNO₃) by radiolysis of dissolved nitrogen
- (4) Contribution due to generation of hydrochloric acid (HCl) by radiolysis of cladding material of the insulation cable (chlorine rubber)
- (5) Contribution due to generation of cesium hydroxide (CsOH) by cesium release in FP
- (6) Contribution by addition of pH adjuster (NaTB) after LOCA

The contribution due to generation of carbonic acid (H₂CO₃) by dissociation of dissolved carbon dioxide was omitted since carbonic acid is a weak acid and the contribution is minor. Figure 06.05.02-1 shows the calculation result of the recirculation water pH variation from the initiation of dissolution of pH adjuster (NaTB) at 3.5 hours after an accident to complete dissolution at 15.5 hours after an accident considering the dissolution delay of pH adjuster (NaTB) to recirculation water.

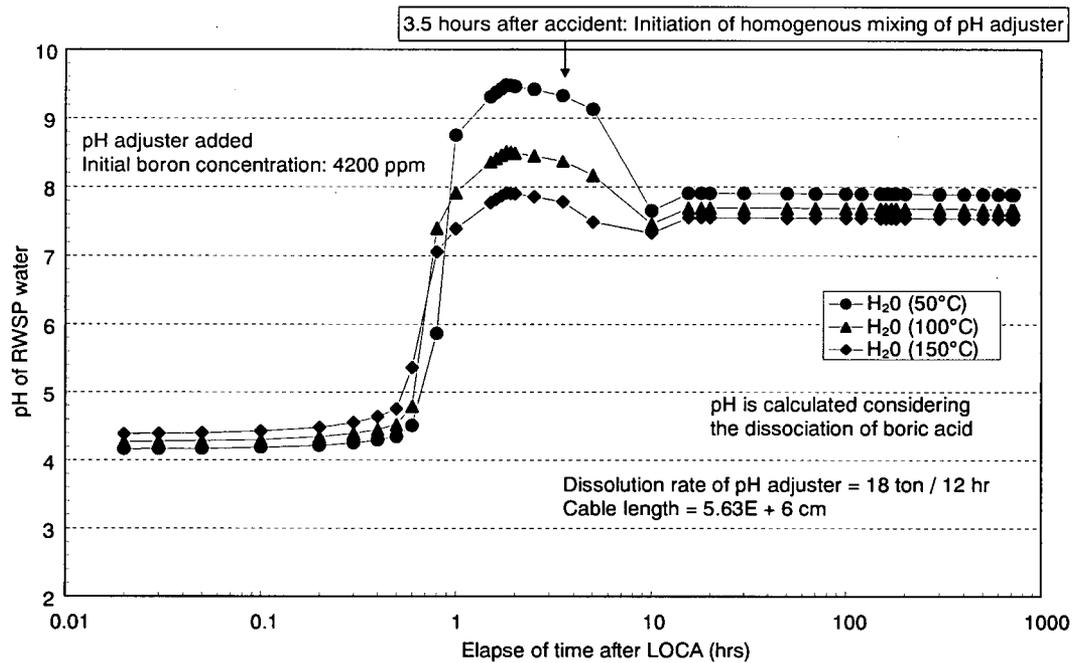


Figure 06.05.02-1 Variation of recirculation water pH with time

According to Figure 06.05.02-1, the recirculation water pH increases with the transition of time after an accident and reaches pH>7 at 0.8 hour after an accident. It is considered that this result was obtained since the contribution by release of alkali (CsOH) was higher than the contribution by (initial) contained boric acid immediately after LOCA and the contribution by generation of acid (HI, HCl, HNO₃) after an accident. The increase margin of pH decreases after that point since it is considered that the contribution by generation of acid (HNO₃, HCl) increases due to radiolysis. The pH gradually decreases to the point around 7 immediately after the addition of NaTB since the dissolution amount is small but pH>7 is maintained after NaTB is completely dissolved.

The pH after the addition of NaTB is evaluated using the expression below.

$$\begin{aligned} \text{pH} &= \text{pKa} + \log [\text{Borate}] / [\text{Acid}] \\ &= \text{pKa} + \log \{ [2y - \text{Hf}] / [2y+x+\text{Hf}] \} \end{aligned} \quad (06.05.02-1)$$

Where, Ka: Dissociation constant of boric acid

[Borate] : Concentration of boric acid ion ($\text{B}(\text{OH})_4^-$)

[Acid] : Concentration of boric acid

x : Initial concentration of boric acid (mols/l)

y : Concentration of pH adjuster per each elapsed time (mols/l)

Hf : Final concentration of hydrogen ion $[\text{H}^+]_{\text{final}}$ of RWSP per unit of elapsed time (mols/l)

The pH evaluation formula of (06.05.02-1) is an approximation expression with a premise that the degree of dissociation of boric acid is low and the borate concentration of the aqueous solution is mostly contributed from the added $\text{Na}_2\text{B}_4\text{O}_7$ (NaTB).

The expression of (06.05.02-1) is almost established herein because the dissociation of boric acid can be ignored when pH is 7 or less. However, the degree of dissociation of boric acid increases as pH increases. Hence, the generation of borate (boric acid ion: $\text{B}(\text{OH})_4^-$) due to the dissociation of boric acid from the pH adjuster and the dissociation of boric acid that was added at the initial stage cannot be ignored. The [Borate] / [Acid] ratio increases and pH becomes higher compared to the value when dissociation of boric acid is ignored.

Thus, the evaluation of pH considering the dissociation of boric acid is described below. The degree of dissociation of boric acid is expressed in the expression below.

$$\begin{aligned} v &= 1 - [\text{Residual boric acid } \text{H}_3\text{BO}_3 \text{ concentration mols/l}] / [\text{Initial boric acid } \text{H}_3\text{BO}_3 \\ &\quad \text{concentration mols/l}] \\ &= [\text{Boric acid ion } (\text{B}(\text{OH})_4^-) \text{ concentration mols/l}] / [\text{Initial boric acid } \text{H}_3\text{BO}_3 \text{ concentration} \\ &\quad \text{mols/l}] \end{aligned} \quad (06.05.02-2)$$

The numerator 2y of the logarithm of the expression (06.05.02-1) is borate (boric acid ion) generated by dissociation of NaTB where 2yv, which is the dissociated portion of boric acid generated by dissociation of NaTB and vX, the dissociated portion of initially added boric acid are added. Thus, the expression is $2y + 2yv + vX = 2y(1 + v) + vX$.

On the other hand, the denominator 2y of the logarithm of the expression (06.05.02-1) is boric acid generated by dissociation of NaTB where the dissociated portion is subtracted and expressed as $2y - 2yv = 2y(1 - v)$. The boric acid added at the initial stage is expressed as $X - vX = X(1 - v)$.

The evaluation expression of pH when the correction of the above is substituted to expression (06.05.02-1) considering the dissociation of boric acid is described below.

$$\text{pH} = \text{pKa} + \log \{ [2y(1 + v) + vX - \text{Hf}] / [2y(1 - v) + X(1 - v) + \text{Hf}] \} \quad (06.05.02-2)$$

The calculation result of Figure 06.05.02-1 shows that the recirculation water pH at the time when the pH adjuster was added (3.5 hours after an accident) is higher than 7, which means that dissociation of boric acid cannot be ignored. Hence, the recirculation water pH of Figure 06.05.02-1 is calculated using the expression (06.05.02-2) considering the dissociation of boric acid.

b. The dissolution rate of NaTB is 3,675 lb/hr (44,100 lb/12hr).

Impact on DCD

There is no impact on the DCD.

Impact on COLA

There is no impact on the COLA.

Impact on PRA

There is no impact on the PRA.

RESPONSE TO REQUEST FOR ADDITIONAL INFORMATION

4/22/2009

**US-APWR Design Certification
Mitsubishi Heavy Industries
Docket No. 52-021**

RAI NO.: NO. 234-2040 REVISION 1
SRP SECTION: 06.05.02 – CONTAINMENT SPRAY AS A FISSION PRODUCT CLEANUP SYSTEM
APPLICATION SECTION: 6.5.2
DATE OF RAI ISSUE: 2/26/2009

QUESTION NO.: 06.05.02-3

- a. How is iodine equilibrium partitioning between liquid and air calculated?
- b. How was possible iodine revolatilization accounted for?
- c. What components are considered in solution?
- d. What pH values are typically obtained for example DBA sequences?
- e. What partition coefficient is used?

Background

GDC 41 establishes the design requirements for containment atmosphere cleanup systems which function to reduce the concentration and quality of fission products released to the environment following postulated accidents. Compliance with GDC 41 ensures that the containment spray system will accomplish the fission product removal function assumed in the Chapter 15 radiological consequence calculations. To meet this design criterion, fission products must be retained as well as removed by the spray solution.

SRP 6.5.2 Acceptance Criteria 1.G. states that long-term iodine retention can only be assumed if sump solution pH remains above 7. Even if most iodine is removed, SRP Review Procedures 4.C.ii recommends an assumption that iodine revolatilization will take place if $\text{pH} < 7$. The analysis that considers the equilibrium partitioning between air and water for I_2 depends strongly on temperature and solution pH. It assumes sufficient mixing by recirculation so that an equilibrium value is a reasonable estimate of the end result of transient dissolution and revaporization processes. SRP Review Procedures 4.C.ii states that the pH calculation must consider all solution components, including fission product oxides, acids generated by radiolysis, and additives. The DCD makes only brief mention of such a calculation, and it is not clear what they would actually calculate (DCD 6.5.2.3.3).

ANSWER:

- a. The partition factor of iodine (= aqueous-phase iodine concentration / gaseous-phase iodine

concentration) at pH of recirculation water after an accident was calculated using the reaction formula and equilibrium constant of iodine and water. Data in literature which covers relatively high temperatures (150°C) (L.F. Parsly, ORNL - TM - 2412 Part IV) was referenced for the equilibrium constant.

The reaction formula and equilibrium constant of iodine and water is described below.

1. $I_2(g) \leftrightarrow I_2(aq)$ $K_1 = I_2(aq) / I_2(g)$
2. $I_2(aq) + I^- \leftrightarrow I_3^-$ $K_2 = I_3^- / I_2(aq) / I^-$
3. $I_2(aq) + H_2O \leftrightarrow H^+ + I^- + HIO$ $K_3 = H^+ \times I^- \times HIO / I_2(aq)$
4. $I_2(aq) + H_2O \leftrightarrow H_2OI^+ + I^-$ $K_4 = H_2OI^+ \times I^- / I_2(aq)$
5. $3I_2(aq) + 3H_2O \leftrightarrow IO_3^- + 5I^- + 6H^+$ $K_5 = (IO_3^-) \times (I^-)^5 \times (H^+)^6 / I_2(aq)^3$

The abovementioned reaction formula 5 becomes significant when pH is 9 or more. Thus, reaction formulas 1 to 4 are considered and 5 is omitted since the neutral region is examined herein.

The expression below is established from the charge balance and proton balance in the solution.

$$6. [HIO] + [H_2OI^+] = [I^-] + [I_3^-]$$

The partition factor H is calculated using the abovementioned expressions 1 to 4 and 6 following the procedure described below.

- a. The expression below is obtained when $[HIO]$, $[H_2OI^+]$, $[I^-]$, $[I_3^-]$ are expressed in functions of $[I_2(aq)]$ and $[H^+]$ from reaction formulas 2 to 4 and 6.

$$[HIO] = \frac{K_3 [I_2]_{aq}}{[H^+] [I^-]}$$

$$[H_2OI^+] = \frac{K_4 [I_2]_{aq}}{[I^-]}$$

$$[I_3^-] = K_2 [I_2]_{aq} [I^-]$$

$$[I^-] = \left[\frac{K_4 [I_2]_{aq}}{1 + K_2 [I_2]_{aq}} + \frac{K_3 [I_2]_{aq}}{[H^+] (1 + K_2 [I_2]_{aq})} \right]^{1/2}$$

- b. The total iodine species concentration in aqueous phase $[I_2]_{aq}(T)$ is derived as a function of $[I_2(aq)]$ and $[H^+]$.

$$7. [I_2]_{aq}(T) = [I_2]_{aq} + 1/2 ([HIO] + [H_2OI^+] + [I^-] + 3 [I_3^-])$$

The analysis result value of total iodine released at LOCA and the value equivalent to the recirculation water pH at the time of an accident at $[H^+]$ was entered in $[I_2]_{aq}(T)$ of expression 7 and $[I_2(aq)]$ was calculated as an unknown quantity.

- c. A comprehensive partition factor H was derived as a function of $[I_2(aq)]$ calculated in expression 7.

$$H = [I_2]_{aq}(T) / [I_2(g)] = K_1 \times [I_2]_{aq}(T) / [I_2(aq)]$$

The calculated partition factor H of iodine when pH is 7 and when the recirculation water pH at LOCA is at the lowest value (final pH after an accident) is described in the table below.

Conditions		50°C	100°C	150°C
pH = 7	pH	7.0	7.0	7.0
	H	1.30E + 3	1.34E + 4	1.09E + 5
At LOCA	pH	7.88	7.67	7.53
	H	9.50E + 3	6.25E + 4	3.70E + 5

b. Because a pH for re-circulating water is greater than 7, iodine revolatilization is not taken into account. (See the response to item d.)

c. Considered components in solution are boric acid, NaTB, I, CsOH, HCl and HNO₃. Concentrations of these components in solution are as following.

Boric acid : 4200 ppm
NaTB : 1.4E-2 mol/l
Iodine : 2.0E-6 mol/l
CsOH : 3.1E-4 mol/l
HCl : 1.1E-3 mol/l
HNO₃ : 5.4E-4 mol/l

(These are values at 30 days with more conservative condition than design value.)

d. The pH variation with time calculated considering factors described below that contribute to the recirculation water pH after an accident is described in aforementioned Figure 06.05.02-1.

- (1) Contribution by (initial) contained boric acid immediately after LOCA
- (2) Contribution due to generation of hydrogen iodide (HI) by iodine release in FP (fission product)
- (3) Contribution due to generation of nitric acid (HNO₃) by radiolysis of dissolved nitrogen
- (4) Contribution due to generation of hydrochloric acid (HCl) by radiolysis of cladding material of the insulation cable (chlorine rubber)
- (5) Contribution due to generation of cesium hydroxide (CsOH) by cesium release in FP
- (6) Contribution by addition of pH adjuster (NaTB) after LOCA

e. Partition coefficient is not used in pH analysis for re-circulating water.

Impact on DCD

There is no impact on the DCD.

Impact on COLA

There is no impact on the COLA.

Impact on PRA

There is no impact on the PRA.