

APPENDIX A

ASSUMPTIONS AND BOUNDARY CONDITIONS

The following list provides the boundary conditions, initial conditions, containment geometry and other assumptions that were used for the parametric studies.

A1. Containment Geometry

Total volume = 50,000 m³

Volume of sump water = 1,000 m³

Liquid-gas interfacial area = 1,000 m²

Surfaces in contact with sump water = 1,000 m²

Surfaces in contact with the gas phase = 25,000 m²

All surfaces are painted ⇒ Each code calculates **organic iodide formation** as a result of the presence of painted surfaces, according to its own mechanism.

All calculations were performed considering the whole containment as a single node.

A2. Initial Conditions and Input Parameters

The initial conditions and input parameters are listed in Tables 7.1 to 7.4.

A3. Output Parameters

1. Total gas-phase iodine concentration, [I(g)], as a function of time.
2. Gas-phase iodine speciation, i.e., [I₂(g)] and [org-I(g)] as a function of time. (If a code allows the further separation of org-I into highly volatile and relatively nonvolatile species, then they can be presented separately. However, always provide the total organic iodide concentration.)
3. Total aqueous-phase iodine concentration, [I(aq)], as a function of time.
4. Aqueous-phase iodine speciation as a function of time: [I₂(aq)], [org-I(aq)] are of main interest, but others such as [IO₃⁻(aq)] and [HOI(aq)], are also welcome.
5. Mass balance of iodine at the end of calculation: percentage of iodine inventory in the gas phase, aqueous phase and on surfaces.

A4. Duration of Calculations

Calculations were performed over a 75-h (3-day) period under a given set of conditions.

A5. Mass Transfer Coefficients

All codes essentially describe a liquid-gas interfacial mass transfer process using a two resistance model. Because of this, and to make the comparison of the results manageable, calculations were performed using same liquid-gas interfacial mass transfer coefficients:

$$\frac{1}{k_{MT}} = \frac{1}{k_{maq}} + \frac{H_{VOLI}}{k_{mg}}$$

$$k_{mg} \text{ (dm}\cdot\text{s}^{-1}\text{)} = 1 \times 10^{-1} (T/298)$$

$$k_{maq} \text{ (dm}\cdot\text{s}^{-1}\text{)} = 7 \times 10^{-3} (T/298)^{2/3}$$

where k_{MT} , k_{mg} and k_{maq} are the overall interfacial, gas-phase and aqueous-phase mass transfer coefficients, respectively, T is temperature in units of K, and H_{VOLI} is the partition coefficient of iodine species VOLI (i.e., I_2 or organic iodides)—see below.

A6. Partition Coefficients

The following partition coefficients were recommended for I_2 , and for high volatile organic iodides (HVRI) and low volatile organic iodides (LVRI):

$$\begin{aligned} \text{For } I_2: & \quad \ln (H_{I_2})_T = \ln 79 + (-3600) \times (1/298 - 1/T) \\ \text{For HVRI (or } CH_3I\text{):} & \quad \ln (H_{HVRI})_T = \ln (4) + (-3400) \times (1/298 - 1/T) \\ \text{For LVRI:} & \quad \ln (H_{LVRI})_T = \ln (1000) + (-6500) \times (1/298 - 1/T)^8 \end{aligned}$$

where T is temperature (K), and $(H_x)_T$ is the partition coefficient of species x at temperature T ; the partition coefficient is defined as the ratio of the aqueous-phase to the gas-phase concentration at equilibrium.

Note that these are only recommended values. Because each code handles organic iodide behaviour differently, the partition coefficients were not fixed for this exercise.

A7. Iodine Adsorption

On Surfaces exposed to the gas phase

⁸ J.C. Wren and G.A. Glowa. Iodine Behaviour Models on Organic Reactions: Partitioning and Hydrolysis/Radiolysis of Organic Iodides,0 (is this “0” correct?). EPRI ACEX Report.
Please contact Jason Chao at EPRI for the report (CHAOJ@epri.com).

The iodine adsorption on dry surfaces (i.e., non-condensing conditions) in contact with the gas phase is described as a first-order process in all models. Calculations were performed using the same deposition velocities for $I_2(g)$ on dry surfaces:

$$\ln (V_{I_2})_T (\text{dm}\cdot\text{s}^{-1}) = \ln (1 \times 10^{-3}) + 5100 \times (1/298 - 1/T)$$

where $(V_{I_2})_T$ is the deposition velocity of I_2 at temperature T in units of $\text{dm}\cdot\text{s}^{-1}$. This equation gives the iodine deposition velocities:

$$\begin{aligned} &1 \times 10^{-3} \text{ dm}\cdot\text{s}^{-1} \text{ at } 25^\circ\text{C}, \\ &2 \times 10^{-2} \text{ dm}\cdot\text{s}^{-1} \text{ at } 90^\circ\text{C}, \text{ and} \\ &3 \times 10^{-2} \text{ dm}\cdot\text{s}^{-1} \text{ at } 100^\circ\text{C}. \end{aligned}$$

On Surfaces exposed to the aqueous phase

For painted surfaces contacting the sump aqueous phase, the values used for the adsorption of I_2 onto (k_{AD}) and the desorption of I_2 from (k_{DES}) were

$$\begin{aligned} k_{AD} (\text{s}^{-1}) &= v_{Paq} \cdot (A_{aq}/V_{aq}) \\ k_{DES} (\text{s}^{-1}) &= 0 \\ \ln(v_{Paq}) &= \ln(v_{Paq(298K)}) + \left(\frac{\Delta E_{vPaq}}{R} \right) \cdot \left(\frac{1}{298} - \frac{1}{T} \right) \\ \Delta E_{vPaq}/R &= 4900 (\text{K}) \\ v_{Paq(298K)} (\text{dm}\cdot\text{s}^{-1}) &= 0.5 \times 10^{-3} \end{aligned}$$

where v_{Paq} is the deposition velocity of $I_2(aq)$ on the surface in contact with the aqueous phase, and A_{aq} is the area of the painted surface in contact with the bulk water phase.

A8. Condensing Conditions

For condensing conditions, all walls were assumed to be covered with condensing water films, and the condensing flow rate, F_{CON} , of $1 \text{ dm}^3\cdot\text{s}^{-1}$ will be used for the calculations:

$$\begin{aligned} F_{CON} &= 1.0 \text{ dm}^3\cdot\text{s}^{-1} \\ \text{Thickness of the water film} &= 5 \times 10^{-4} \text{ m} \\ V_{CON} &= 12.5 \text{ m}^3 \end{aligned}$$

where V_{CON} is the total volume of condensate.

A9. pH Profile for Case 4

For the set of calculations (see Case 4 in the tables) for which pH varies with time, the following pH profile was used:

$$\text{pH} = -3 \ln((t + 1)^{1/2}) + 10$$

where t is time in **hours**.

This equation provides the pH profile shown in Figure A1.

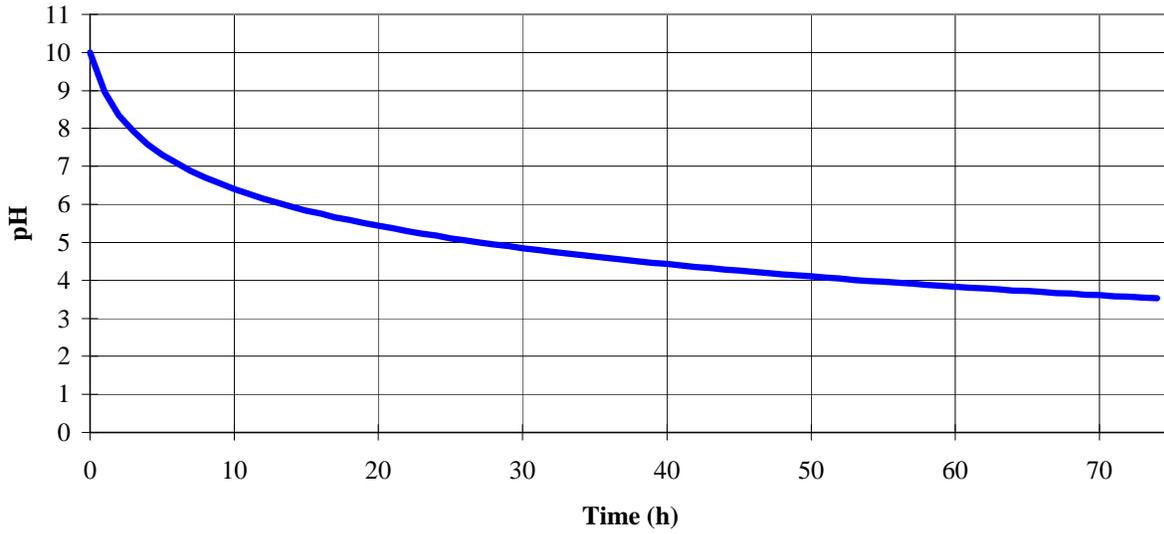


Figure A1. pH profile to be used for the calculations under case 4 conditions.

TABLES

CONDITIONS USED FOR THE PARAMETRIC STUDIES

TABLE A.1. No silver, non-condensing (i.e., no wet surfaces, except those in contact with the sump water).

Case 1: Effect of temperature, constant pH, # of sets of calculations = 12				
Case #	Temperature (°C)	pH	Initial $[\Gamma]_0$ (mol·dm ⁻³)	Dose rate (aq) (kGy·h ⁻¹)
1A9, 1A7, 1A5, 1A4	90	9,7,5,4	1×10^{-5}	1
1B9, 1B7, 1B5, 1B4	130	9,7,5,4	1×10^{-5}	1
1C9, 1C7, 1C5, 1C4	60	9,7,5,4	1×10^{-5}	1
Case 2: Effect of initial $[\Gamma]_0$, constant pH, # of sets of calculations = 8				
Case #	Temperature (°C)	pH	Initial $[\Gamma]_0$ (mol·dm ⁻³)	Dose rate (aq) (kGy·h ⁻¹)
2A9, 2A7, 2A5, 2A4	90	9,7,5,4	1×10^{-6}	1
2B9, 2B7, 2B5, 2B4	90	9,7,5,4	1×10^{-4}	1
Case 3: Effect of dose rate (aq), constant pH, # of sets of calculations = 8				
Case #	Temperature (°C)	pH	Initial $[\Gamma]_0$ (mol·dm ⁻³)	Dose rate (aq) (kGy·h ⁻¹)
3A9, 3A7, 3A5, 3A4	90	9,7,5,4	1×10^{-5}	10
3B9, 3B7, 3B5, 3B4	90	9,7,5,4	1×10^{-5}	0.1
Case 4: Varying pH, # of sets of calculations = 3 pH = 3 ln ((t + 1)^{1/2}) + 10, t in hours				
Case #	Temperature (°C)	pH	Initial $[\Gamma]_0$ (mol·dm ⁻³)	Dose rate (aq) (kGy·h ⁻¹)
4A	90	varying with t	1×10^{-5}	1
4B	90	varying with t	1×10^{-4}	1
4C	90	varying with t	1×10^{-6}	1
Case 5^a: Uncontrolled pH, starting pH = 10, # of sets of calculations = 3 Comparison of different acid formation mechanisms used in various codes.				
Case #	Temperature (°C)	pH	Initial $[\Gamma]_0$ (mol·dm ⁻³)	Dose rate (aq) (kGy·h ⁻¹)
5A	90	uncontrolled	1×10^{-5}	1
5B	90	uncontrolled	1×10^{-5}	10
5C	60	uncontrolled	1×10^{-5}	1

^aCase 5 is not required for codes for which pH is an input parameter. (continued)

TABLE A.2. No silver, condensing (condensate volume = 12,500 dm³, volume flow rate = 1 dm³/s, condensing film thickness = 5×10⁻³ dm).

Case 6: Effect of condensation, constant pH, # of sets of calculations = 8				
Case #	Temperature (°C)	pH	Initial [I] _o (mol·dm ⁻³)	Dose rate (aq) (kGy·h ⁻¹)
6A9, 6A7, 6A5, 6A4	90	9,7,5,4	1 × 10 ⁻⁵	1
6B9, 6B7, 6B5, 6B4	130	9,7,5,4	1 × 10 ⁻⁵	1

TABLE A.3: Non-condensing, silver present (amount of Ag = 100 kg in the sump water, surface area of Ag = 800 m²/kg Ag, 10% of Ag is present as AgO_x for a code which requires oxidized silver).

Case 7: Effect of silver, constant pH, # of sets of calculations = 8				
Case #	Temperature (°C)	pH	Initial [I] _o (mol·dm ⁻³)	Dose rate (aq) (kGy·h ⁻¹)
7A9, 7A7, 7A5, 7A4	90	9,7,5,4	1 × 10 ⁻⁴	1
7B9, 7B7, 7B5, 7B4	90	9,7,5,4	1 × 10 ⁻⁵	1

TABLE A.4. No silver, non-condensing, organic impurities initially present in the sump.

(For these calculations, no other sources of organic impurities in the sump water, such as the dissolution of solvents from painted surfaces, or no organic iodide formation other than via reactions with the initially present organic impurities, would be considered.)

Case 8: Constant pH, # of sets of calculations = 12				
Effect of organic impurities in the sump:				
for A & B, [Org]_o = 1×10⁻³ mol·dm⁻³; for C, [Org]_o = 1×10⁻⁵ mol·dm⁻³.				
Case #	Temperature (°C)	pH	Initial [I] _o (mol·dm ⁻³)	Dose rate (aq) (kGy·h ⁻¹)
8A9, 8A7, 8A5, 8A4	90	9,7,5,4	1 × 10 ⁻⁵	1
8B9, 8B7, 8B5, 8B4	130	9,7,5,4	1 × 10 ⁻⁵	1
8C9, 8C7, 8C5, 8C4	90	9,7,5,4	1 × 10 ⁻⁵	1