Technical Assistance in Review of Source Term–Related Issues of Advanced Reactors

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Abstract

The distribution of iodine in containment during an AP-600 design-basis accident was evaluated using models in the "TRENDS" code. The AP-600 3BE accident sequence calculations showed that a pH >7 was maintained for at least 30 days. Because the pH was maintained at this level, most of the iodine was in the form of iodide; only 3×10^{-3} % was present as aqueous I_2, and only 1×10^{-6} % was present as I_2 in the vapor phase.

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Executive Summary

Results of previous studies have shown that pH is the major factor in determining the amount of I_2 and organic iodides in containment water pools. When the pH is <7 (acidic), these volatile iodine species can form. The major contributors to acidity are the acids (hydrochloric and sulfuric) produced by the irradiation and/or heating of Hypalon™ (a registered trademark of DuPont for chlorosulfonated polyethylene rubbers) cable jacketing material.

The distribution of iodine in containment during an AP-600 design-basis accident was evaluated using a group of models known collectively as the "TRENDS" code. In this study, the 3BE sequence (a medium loss-of-coolant accident) was examined. In this analysis, thermal hydraulic data were inserted into TRENDS models along with physical and chemical parameters specific to AP-600 containment. The calculations were performed for times up to 24 h into the accident sequence, based on thermal hydraulic data from the MAAP code. These data were extrapolated to permit an evaluation of iodine behavior for a 30-day period.

The AP-600 3BE accident sequence calculations showed that a pH >7 was maintained for at least 30 days. Approximately 360 mol of hydrochloric acid from Hypalon™ jacketing was indicated in the calculations. Because the pH was maintained above 7, most of the iodine was in the form of iodide in water pools and was not present in a volatile form such as I_2 or an organic iodide. The overall iodine distribution was aqueous iodide (>99.9%), aqueous I_2 (3×10^{-3}%), and I_2 in the vapor phase (1×10^{-6}%). Iodide deposited on surfaces was not considered.

1 Introduction

In a light water reactor (LWR) accident sequence, fission products released from the core will undergo changes in temperature and concentration as they pass through regions of the reactor coolant system (RCS) and enter containment. A study of seven accident sequences[1] showed that iodine entering the containment from the RCS was almost entirely in the form of cesium iodide (CsI). The chemical forms of iodine entering containment were allocated as 95% CsI and a maximum of 5% as elemental iodine (I_2) and HI.[1] Iodine in the form of I_2, HI, or some small organic iodides such as methyl iodide (CH_3I) or ethyl iodide (C_2H_5I), is volatile even at room temperature. This means that each of these species can be transported in the vapor phase.

Once the fission products are in containment, pH will be the major factor in determining iodine chemical forms.[2] Two chemical processes are involved here: hydrolysis and radiolysis. In hydrolysis, I_2 is converted to iodide (I^-) and iodate (IO_3^-) at pH values >7. In radiolysis, I^- is converted to I_2 at pH values <7. Both processes occur in irradiated water pools; and at a given temperature, the net result depends on pH and radiation dose. In addition, the major mechanism for forming organic iodides involves the combination of radiation, pH values <7 that lead to the formation of I_2, and organic material. Thus, pH is the dominant factor in establishing all iodine chemical forms in containment; and acid pH values lead to forming and maintaining I_2, as well as to the formation of organic iodides.

In containment, the materials that will determine pH include boric acid, nitric acid from irradiated water pools, hydrochloric acid (HCl) and sulfuric acid (H_2SO_4) from irradiated and/or heated cable jacketing, basic fission product compounds such as cesium hydroxide or cesium borate, core-concrete aerosols, and basic pH additives such as sodium hydroxide or sodium phosphate. In a previous study, *Iodine Evaluation and pH Control* (NUREG/CR-5950),[2] it was shown that acid from the irradiation and heating of Hypalon™ (a registered trademark of DuPont for chlorosulfonated polyethylene rubbers) cable jacketing could bring about acidic pH values if sufficient basic additives are not available.

The primary concern in iodine studies is the distribution of iodine in containment—especially the fraction of iodine that is in the vapor phase. The evaluation of iodine distribution requires thermal hydraulic data for plant-specific sequences and an array of models that permit calculation of parameters such as radiation dose rates, pH values, iodine chemical forms, and transport of iodine from the aqueous phase to the vapor phase. The models, described in Reference 3, must be specially assembled for each containment to ensure that the control volumes and materials are represented properly. This effort requires the coupling of thermal hydraulic data for an accident sequence with models and applying this information to the physical and chemical characteristics of a specific containment. The distribution of iodine in containment during a loss-of-coolant accident (LOCA) in an AP-600 system is described in Section 2.

2 Distribution of Iodine Species in Containment During an AP-600 Design-Basis Accident

The distribution of iodine in postulated reactor accidents has been a difficult, but important, issue for many years. It is difficult because the behavior of iodine can vary over a wide range. Some species are highly volatile, while others are stable and fixed. It is important because iodine would pose the major short-term threat to human health and safety in the event of an accident.

Standard reactor analysis codes, such as MELCOR and MAAP, are not equipped to evaluate the transient behavior of iodine since they assume that it always appears in the aerosol form CsI. While this may often be the case, some conditions encountered during accidents might favor the formation of I_2 or organic iodides, which are much more volatile. Since conditions vary from plant to plant and sequence to sequence, each postulated accident must be evaluated individually to determine the likely forms, and subsequent volatilization, of iodine.

Under Nuclear Regulatory Commission (NRC) guidance, models of iodine behavior under various severe accident conditions have been developed at Oak Ridge National Laboratory (ORNL). These models have come to be known collectively as the "TRENDS" code. NRC support of model development ended in 1992, before any formal documentation or code development was undertaken; however, at that time, a need arose to actually apply most of the iodine chemistry and interaction models to

postulated severe accident sequences at the High Flux Isotope Reactor (HFIR) in Oak Ridge. As a part of this work, substantial documentation was accomplished.[3] In addition, the HFIR-TRENDS code was developed to actually perform the calculations. This code is not easily exportable or transportable to other plants or sequences, inasmuch as many statements are tailored to the particular plant (i.e., HFIR) for which they were written. However, in the current project, certain parts of HFIR-TRENDS have been appropriated and modified so as to simulate two important features of the AP-600 design-basis accident: (1) the changing pH in various water volumes, and (2) the effect the changing pH might have on the aqueous conversion of iodide to more volatile iodine species.

The accident selected for analysis (by Westinghouse and NRC personnel) is the 3BE accident sequence, which represents a medium LOCA, with a double-ended guillotine break of primary recirculation piping in steam generator (SG) room 2. In addition, a failure of gravity drain injection to the core is assumed; In-containment Refueling Water Storage Tank (IRWST) flow is initiated by operator action. According to the AP-600 PRA, Rev. 1, this accident would be the greatest contributor to the total core damage frequency.

As requested by Westinghouse, a design-basis accident was constructed by using transient conditions for the 3BE sequence, truncated to avoid vessel failure. Release rates for all nuclide groups, as specified in Reference 4 and acid generation rates from Reference 2, were imposed on the resulting thermal-hydraulic behavior. Since the focus of this study was on pH control and aqueous speciation of iodine, fission product iodine was input as CsI into containment water; hence, no aerosol dynamics were modeled.

The TRENDS models[3] do not include primary system, aerosol, or thermal hydraulic analyses. The basic plant analysis was done by Westinghouse using the MAAP code. Transient variables such as temperature, pressure, and fission product inventories were then input into the TRENDS analysis. The control volumes listed in Table 2.1 and the flow paths shown in Figure 2.1 are exactly those of the MAAP calculation. Other than iodine inventories, all values obtained from MAAP were assumed to be unaffected by pH or iodine speciation.

Table 2.1 Identification of control volumes

1	Steam generator room 1
2	Steam generator room 2
3	Lower compartment
4	Cavity
5	IRWST room
6	Upper-compartment cylinder
7	Upper-compartment annulus
8	Upper-compartment dome
9	Valve vault

All unidirectional flows calculated by the MAAP code were used to calculate exchanges between various control volumes. However, the countercurrent (i.e., buoyancy-driven) flows were not used since NRC has yet to accept the validity of this modeling approach. Thus, the mixing of water and gases calculated in this study should differ substantially from calculations obtained by MAAP.

The calculation of pH is performed in each control volume at each time step, immediately prior to the iodine speciation calculation. It assumes that the system is constantly in chemical equilibrium with respect to species that significantly affect pH. Internal coding ensures that time steps are small, so that changes in chemical inventories of each control volume are also small.

The actual equilibrium computation is performed using the principal subroutines of the SOLGASMIX code, as described in Reference 3.

ORNL DWG 95A-744
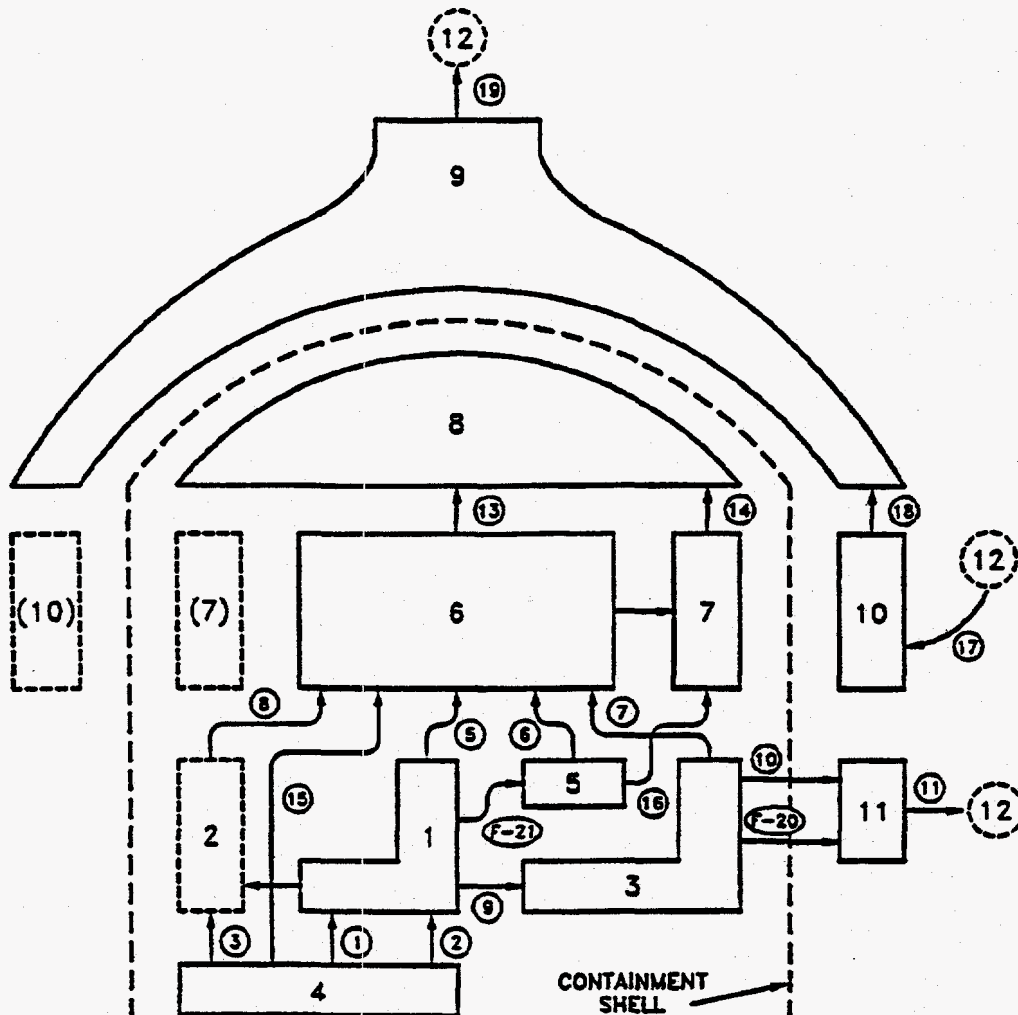
Figure 2.1 MAAP 4.0 description and AP-600 modeling, nodes, and flow paths

In any reactor system, pH values in sumps and pools will depend on the following considerations:

(1) chemicals normally in the reactor system, such as borates, NaOH, or Na_3PO_4;
(2) radiolytic formation of nitric acid in air/water systems;
(3) radiolytic or thermal formation of HCl from cable sheathing;
(4) fission product CsOH; and
(5) radiolysis and hydrolysis of iodine.

The particular assumptions in this application are as follows:

(1) All water from the RCS has a boron concentration of 0.25 mol/L (2700 ppm). This includes normal RCS water, pressurizer, accumulators, core makeup tank (CMT), etc.

Distribution of Iodine

(2)   The IRWST water has a boron concentration of 0.25 mol/L (2700 ppm).

(3) HNO_3 formation is modeled according to Reference 2; the total amount formed is <10 mol, which is not significant.

(4)   HCl formation is modeled according to Reference 2.

(5) Na_3PO_4 is dissolved on the basis of the water height in SG room 1 (it should be recalled that the pipe break occurs in SG room 2), so that all phosphate is in solution when a height of 3 m (9.8 ft) occurs. The total phosphate present was assumed to be 9309 mol Na_3PO_4 (7830 lb, of which 42.97% is anhydrous Na_3PO_4).

Several of the models depend on the prediction of decay energy in air or water. Energy output values for beta and gamma radiation are given in Table 2.2 for each fission product group at various times. These values were calculated using ORIGEN[4] results for fission product inventories, together with ICRP data[5] for nuclide decay energies.

Table 2.2   Radiation energy to water pool

Beta-energy output by MAAP fission product group [(MeV/s)/kg $\times 10^{-12}$]

Time (h)	Xe 1	CsI 2	TeO$_2$ 3	SrO 4	MoO$_2$ 5	CsOH 6	BaO 7	La$_2$O$_3$ 8	CeO$_2$ 9	Sb 10	Te$_2$ 11	UO$_2$ 12
0	12.470	230.89	134.70	38.138	10.040	34.251	69.500	57.823	8.365	650.10	168.00	0.337
1	5.946	185.28	53.26	35.527	4.500	17.302	29.253	34.856	8.292	232.70	66.43	0.169
2	4.729	152.24	29.82	33.271	4.134	9.956	19.828	30.953	8.186	141.80	37.19	0.138
5	3.481	109.61	15.39	28.209	3.987	4.114	11.146	25.192	7.881	87.83	19.20	0.128
10	2.883	88.071	11.891	23.071	3.782	1.564	8.876	19.865	7.405	56.39	14.83	0.120
24	2.068	61.725	8.980	16.831	3.266	0.549	8.418	14.752	6.300	31.67	11.20	0.101
48	1.362	41.878	7.093	13.990	2.537	0.512	7.985	13.030	5.002	24.700	8.846	0.076
96	0.954	24.966	4.869	13.117	1.533	0.505	7.184	11.974	3.698	17.820	6.073	0.043

Gamma-energy output by MAAP fission product group [(MeV/s)/kg $\times 10^{-12}$]

Time (h)	Xe 1	CsI 2	TeO$_2$ 3	SrO 4	MoO$_2$ 5	CsOH 6	BaO 7	La$_2$O$_3$ 8	CeO$_2$ 9	Sb 10	Te$_2$ 11	UO$_2$ 12
0	19.470	753.32	235.01	60.098	15.169	52.232	56.141	41.486	4.597	2253.0	293.10	0.119
1	8.780	615.56	97.90	49.627	2.386	21.006	10.097	27.007	4.549	793.10	122.10	0.097
2	6.729	500.61	54.26	41.258	1.620	9.718	7.156	22.746	4.481	466.50	67.67	0.092
5	4.005	347.38	25.68	24.911	1.525	4.518	6.473	17.590	4.280	270.70	32.03	0.088
10	2.298	278.13	21.53	12.608	1.447	3.654	6.317	15.348	3.971	161.00	26.85	0.083
24	1.084	199.53	17.62	3.498	1.249	3.317	6.153	14.045	3.255	77.95	21.98	0.070
48	0.529	144.10	12.98	0.592	0.970	3.257	5.900	13.243	2.418	59.510	16.190	0.052
96	0.317	91.37	7.51	0.018	0.586	3.164	5.432	12.067	1.587	44.590	9.368	0.030

Doses to water assume that all energy from beta and gamma decay is absorbed in water. They were calculated by combining information from Table 2.2 (interpolated to the correct time) with the water inventories of each fission product group at each time step, as obtained from MAAP output.

The rate of HCl formation in cable sheathing is based on the energy production rates (both beta and gamma) due to fission products that are airborne or deposited on surfaces. The dose rates, which were obtained from Westinghouse, are shown in Figure 2.2. These are similar (for both beta and gamma) to our own calculations using only noble gases and airborne iodine for dose rates in containment airspace. The Westinghouse values also allow for contributions from deposited aerosols. The HCl production is calculated from this total decay energy by using the procedure described in Reference 2. (Note the difference between energy produced and dose absorbed.) The cumulative HCl produced is shown in Figure 2.3 for the first day.
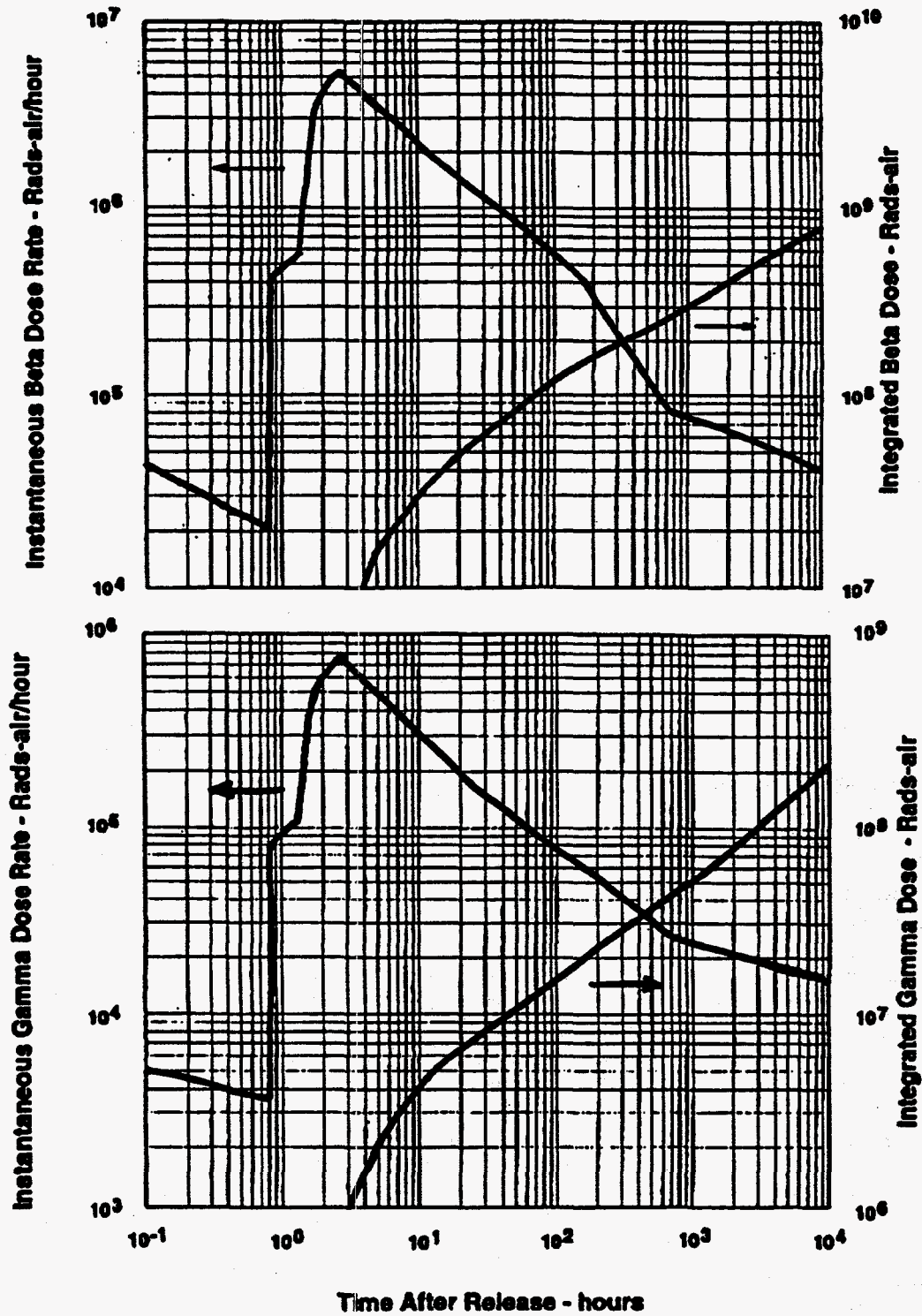
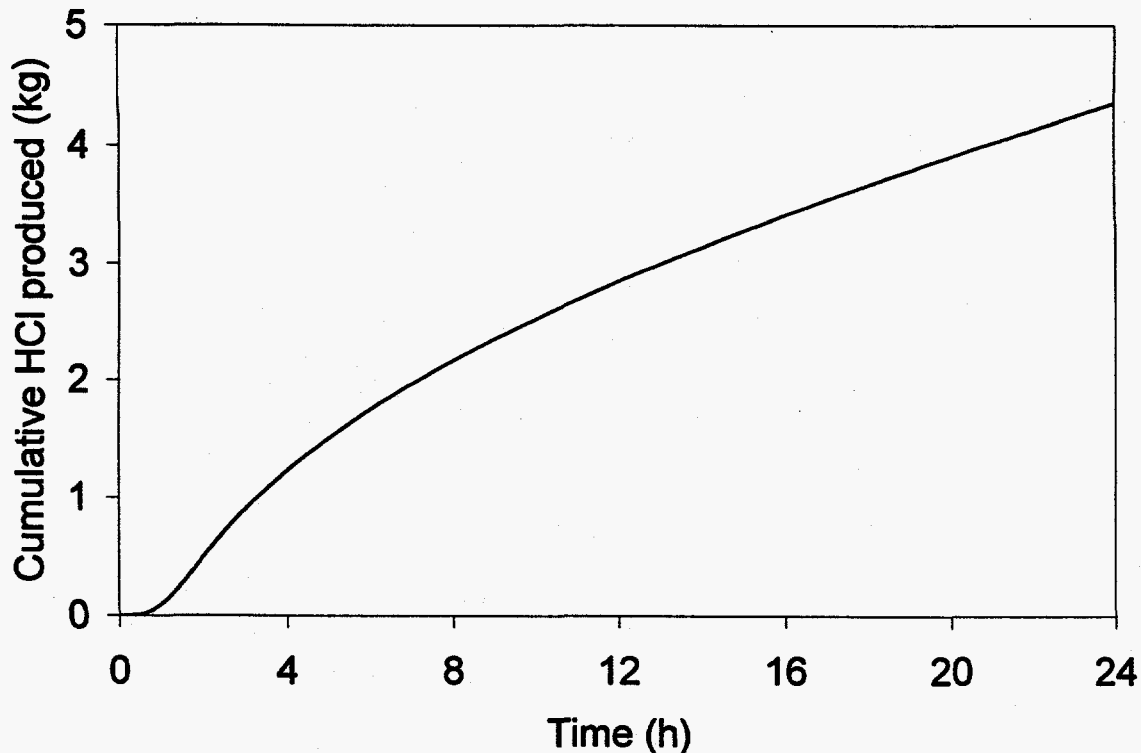Figure 2.2   Containment dose rates

5

Figure 2.3   Acid production from cable insulation

Fission products were sourced into SG room 2, where the hypothesized pipe break occurred. Noble gases were input to containment air and, together with any volatile iodine, constitute the radiation dose to airspace and cable insulation. All other fission products were input to water and were expected to move with the water (i.e., no settling, deposition, or holdup was considered). The release rates, which were taken from Reference 6, encompass a gap release beginning at accident initiation and lasting 0.5 h, followed immediately by an in-vessel release (duration, 1.3 h). The release rates for each of the fission product groups are shown in Table 2.3. The last column notes the energy deposition rate for the total group source (including gap and in-vessel releases). The in-vessel fraction for the ruthenium group was estimated, since the nuclides in this group do not correspond exactly to a single nuclide group in Reference 6. The calculation of this value is discussed in Appendix A.

The alleged pipe break occurs in SG room 2, but the liquid volume increases more rapidly in SG room 1 and in the cavity. Evidently, the direct flows do not account for all of this; hence, the input of fission product sources into the SG-2 volume creates a high concentration (several molal) for the first several time steps. This gradually dissipates as these three volumes mix. In order to overcome this anomaly, fission products were input into all three volumes, according to the liquid volume in each. Total source rates into containment for noble gases, iodine, and nonvolatile fission products, are shown in Figure 2.4.

Trisodium phosphate was sourced into the SG-1 volume, as shown in Figure 2.4. The source rate depended on the water level in this volume, as calculated by MAAP, so that when the water level reached 3 m (9.84 ft), all phosphate was assumed to be in solution. As shown in Figure 2.4, a small fraction of the dissolution occurs very quickly due to the small water volume, which represents wall condensation. However, most of the phosphate remains undissolved until the bulk water level rises sufficiently (at approximately 1.5 h). The initial phosphate dissolution, while small, has a significant effect on pH and, therefore, on iodine retention. Without it, fission products would be released directly into hot, borated water (pH ≤ 5.5), and more volatilization would undoubtedly occur. The initial phosphate source helps alleviate this effect, and the pH is well controlled once the bulk of the phosphate has been dissolved. It is possible that this effect is an anomaly, and that no phosphate would enter solution until the bulk level rises.

Table 2.3 Release rates of primary fission product groups into containment

AP-600 fission product group inventories

| Fission product group		2-h Inventory (g)	Gap release, 0.5-h duration	Fractions[a] in-vessel, 1.3-h duration	Rate of mass addition (g/s)		Total energy deposition rate (MeV/s) × 10^18
					Gap	In-vessel	
1	Xe	411,900	0.05	0.95	11.4417	83.6122	4.719
2	I	18,360	0.05	0.35	0.51	1.3731	9.604
3	Cs	237,600	0.05	0.25	6.6	12.6923	1.590
4	Te	34,250	0	0.05	0	0.3659	0.180
5	Sr	70,700	0	0.02	0	0.3021	0.124
6	Mo	243,600	0	0.0025	0	0.1301	0.005
7	Ba	107,800	0	0.02	0	0.4607	0.065
8	La	566,200	0	0.0002	0	0.0242	0.007
9	Ce	200,800	0	0.0005	0	0.0215	0.002
10	Sb	2,037	0	0.05	0	0.0218	0.062
11	U	64,360,000	0	0.0005	0	6.8761	0.009
12	Ru	612,000	0	0.0009	0	0.1177	0.018
				Total nonvolatile (groups 4–12)		8.320	0.472

[a]Adapted from Reference 4.
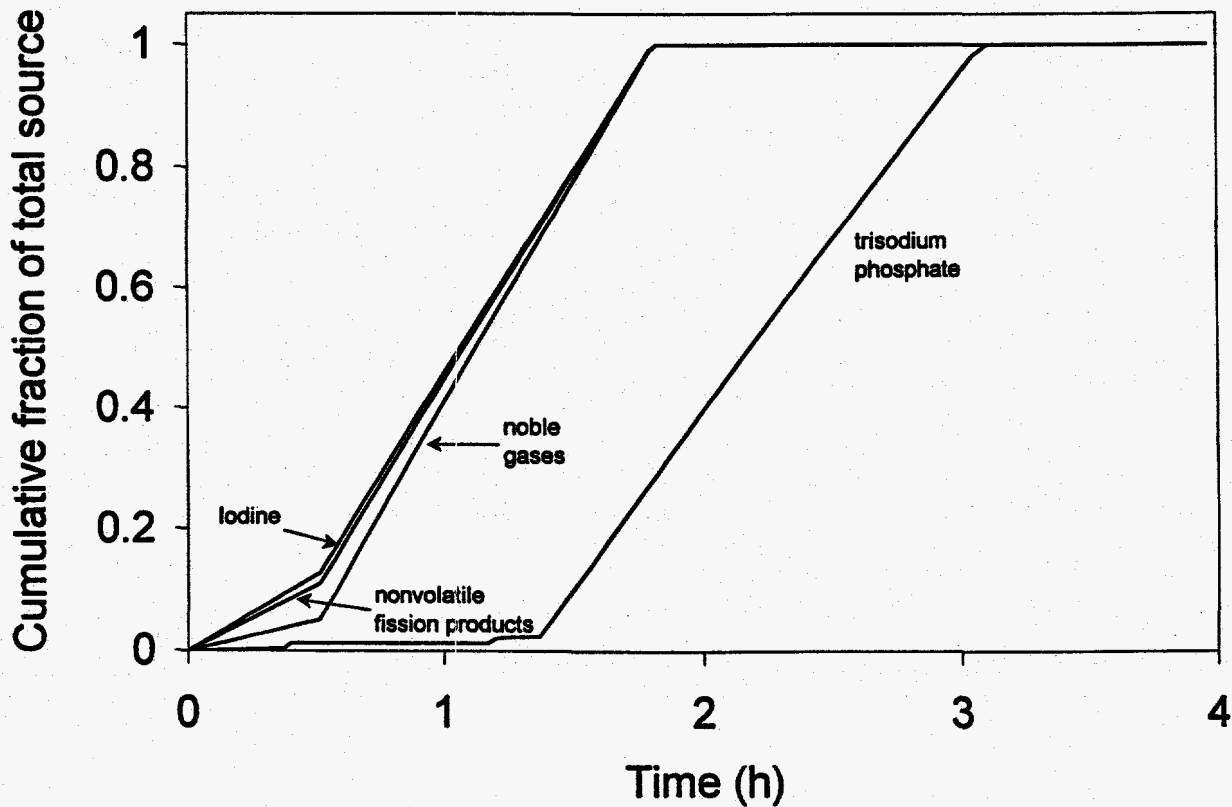
Figure 2.4 Sources and additions into containment

2.1 Twenty-Four-Hour Analysis

Most of the thermal-hydraulic output from the MAAP code is available only for the first 25 h of the accident sequence. Hence, the pH and the iodine speciation analysis are based directly on this computed description of the accident.

Transient pH levels are shown in Figures 2.5–2.7 throughout the first day, grouped according to similarities in behavior. No iodine enters volumes 5, 7, or 8; thus, the low pH values are of little significance to iodine volatility (cf. Figure 2.7). The pH calculations were benchmarked by laboratory measurements at the calculation temperature. In one case at 77.3°C, the calculated pH was 4.57 and the measured pH was 4.63. In another case at 63°C, both the calculated and measured pH values were 7.72. Because of the flow anomalies mentioned earlier, aqueous fission products enter only control volumes 1, 2, and 4 during the first 10 h. As seen in Figure 2.5, the pH in these volumes is at or above neutral for the duration of the sequence (at least during the first day). The near-neutral conditions initially in volumes 2 and 4 contribute to a small amount of volatilization; however, this is not sustained since mixing with high-phosphate water increases the pH to safe levels within 2 h.

For several hours, control volumes 3, 6, and 9 contain no fission products or phosphate. The water present is mostly that from condensation; hence the pH is quite low, as shown in Figure 2.6. However, as the containment water level continues to rise, bulk water does enter these volumes, bringing both fission products and phosphate. This accounts for the rapid rises in pH shown in Figure 2.6. However, because iodine and phosphate enter simultaneously, no significant volatilization occurs.

The iodine distribution in the entire containment is shown in Figure 2.8. The designation I⁻ (aq) refers to dissolved ionic species. This is >99% iodide, I⁻, with a small contribution from iodate, IO_3^-. Similarly, I_2 (aq) refers to several dissolved molecular species, including I_2 (aq), HOI, HIO_2, and CH_3I. The symbol I_2 (g) represents the combination of I_2 and CH_3I in the gas phase, with >99% as I_2. Because of the small volatilization, no surface deposition was calculated.

2.2 Thirty-Day Analysis

Whereas the 24-h analysis was based on containment conditions calculated by the MAAP code, such computations do not exist for the remainder of the 30-day period. Hence, considerable approximations and assumptions are necessary to envision a description of containment behavior during this time interval.

All principal aqueous repositories were combined (i.e., control volumes 1, 2, 3, 4, and 6), which is a reasonable depiction of the actual containment conditions. The flooded containment is really one body of water, although it occupies several rooms and compartments. While previously defined as separate control volumes, there is considerable interconnection and circulation between them. (The warmest region—the reactor core and reactor cavity—is also the lowest, creating effective natural circulation.) Because this flow is unquantified and temperature differences in various regions are not excessive, it is reasonable to assume a single well-mixed control volume for the 30-day period. Control volumes 7 and 8 are also included and contribute the principal gas space for the containment as a whole.

Control volume 5 (the IRWST) is completely isolated from the rest of containment, since no connecting flow occurs and no fission products are present; hence, it is not included in the 30-day analysis. In addition, volume 9 (valve vault) is not included. While there is a small fission product inventory and a slowly increasing water volume (due to seepage from the other containment volumes), this volume is isolated from the environment. Any volatile iodine would have to reenter the principal containment volume before being released to the environment. Since the amount would be quite small, its behavior would ultimately be determined by conditions in the main containment volume.

We thus consider one large volume composed of liquid and gaseous inventories from volumes 1, 2, 3, 4, 6, 7, and 8 at the end of the 24-h calculation. Total system pressure is given below for the last half of the first day.

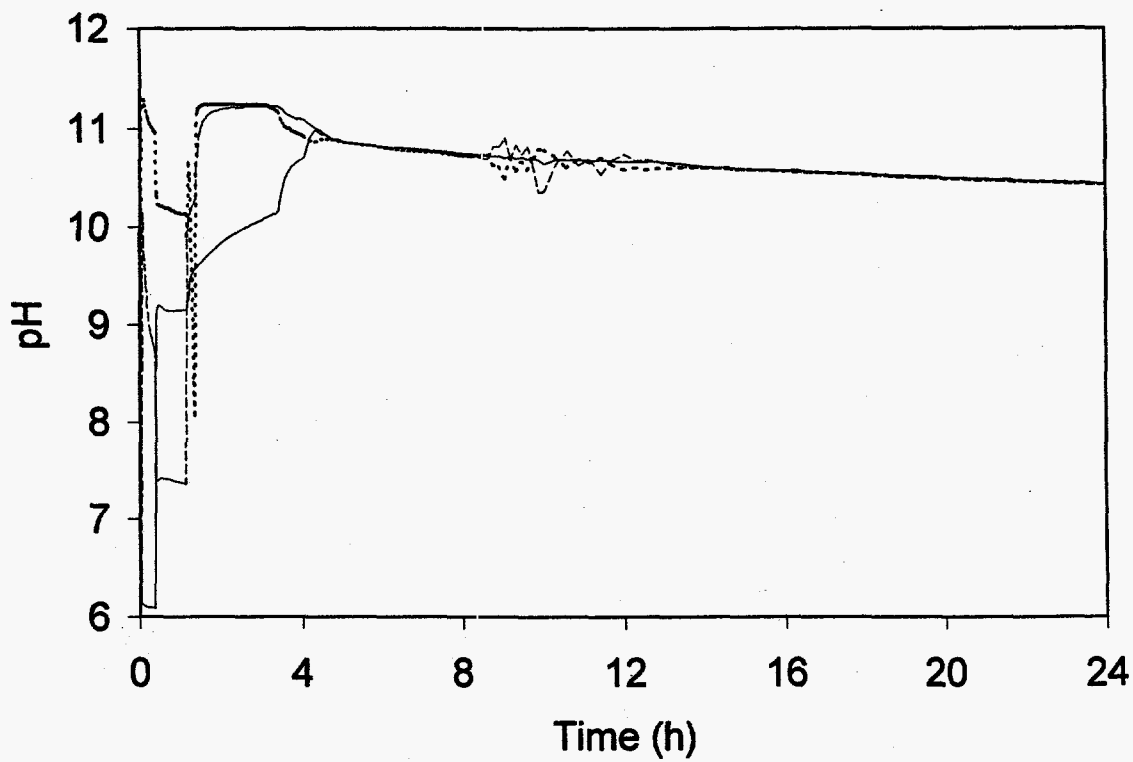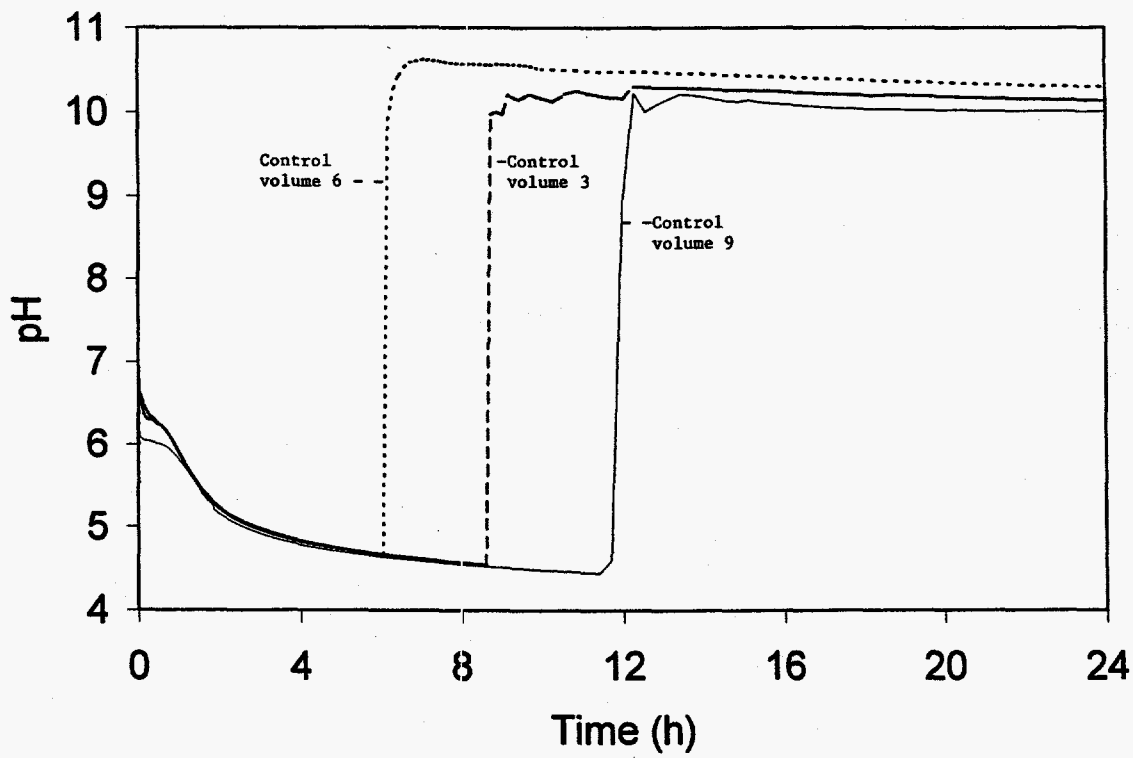| Time (h)	16	18	20	22	24
Pressure (atm)	1.79	1.78	1.76	1.76	1.75

Figure 2.5 pH in control volumes 1, 2, and 4

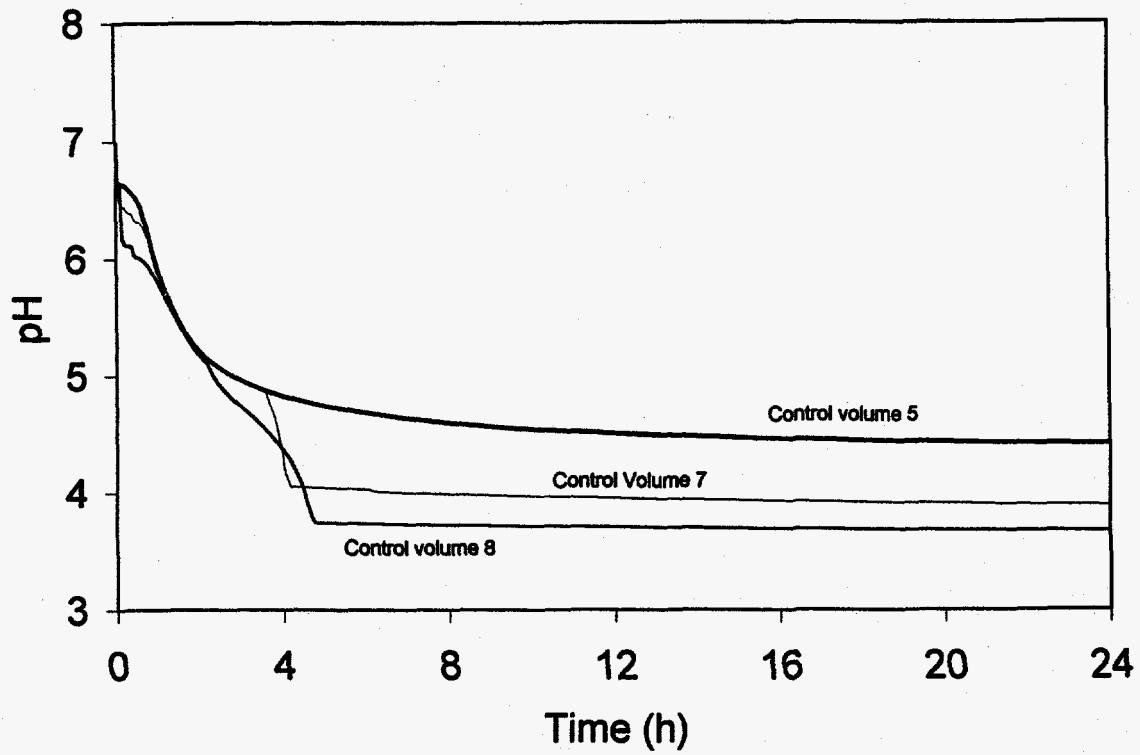Figure 2.6 pH in control volumes 3, 6, and 9

9

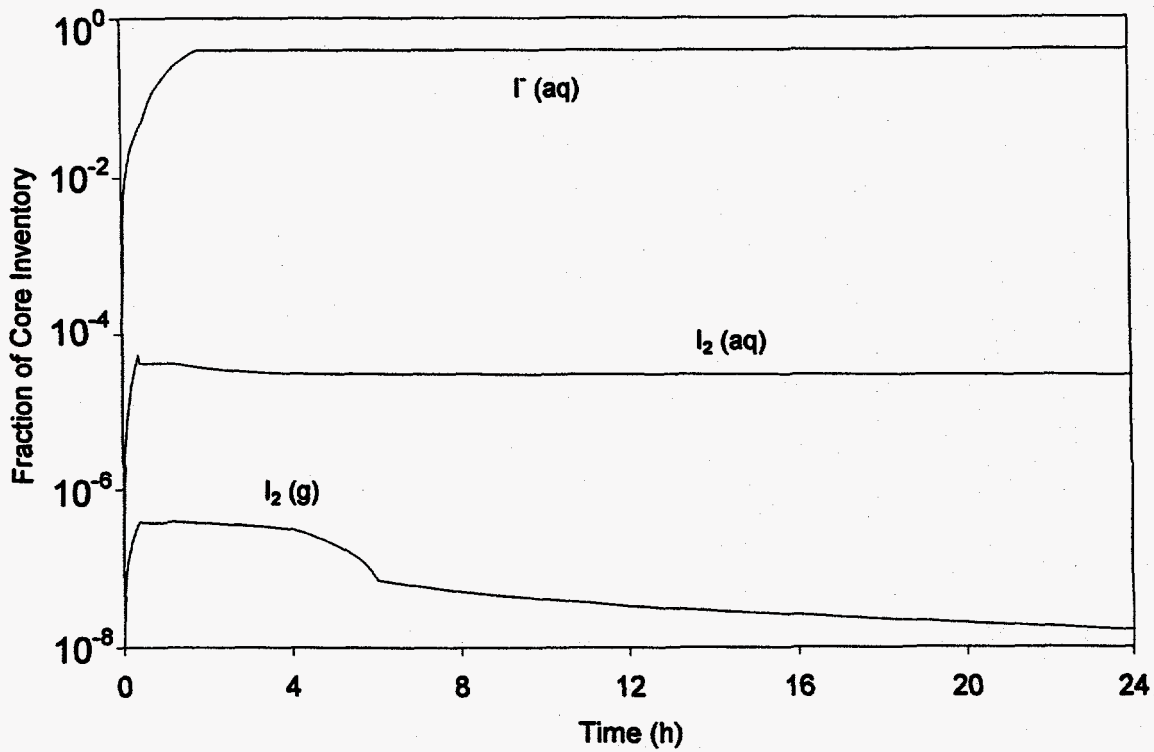**Figure 2.7   pH in control volumes 5, 7, and 8**

Figure 2.8   Total iodine in containment

We assume that the pressure stays constant at 1.75 atm for the duration of the 30-day period, and that bulk gas and liquid temperatures are at saturation for pure water [i.e., 389.3 K (241.14°F/116.2°C)], which is a reasonable average of gas and liquid temperatures at the end of the first day. Of course, in an actual accident, we would not expect constant bulk temperatures in either the gas or the liquid but, instead, strong natural convection driven by temperature gradients. However, we are not seeking to evaluate natural convection; we assume it exists and creates well-mixed volumes. Our purpose is to evaluate water pH and iodine chemistry, for which the bulk average should suffice.

The only truly dynamic behavior is that which would impact water chemistry. The initial borate concentration does not change. All phosphate is in solution, as are any fission products to be considered. Both water and gas volumes and temperatures are established. (Even if not constant in reality, they would probably not change enough to significantly affect water chemistry.)

The single nonconstant effect is the continued generation of acid due to the radiolysis of cable insulation and other materials. This phenomenon continues to be modeled as it was in the 24-h case (i.e., using Westinghouse dose estimates from Figure 2.2). This dose rate, which produces acid according to the empirical model in Reference 2, is shown for the 30-day period in Figure 2.9. This production rate decreases over time, due to decreasing dose rates (cf. Table 2.2). As seen in Figure 2.10, the acid production does result in a small decrease in pH, but this is negligible due to the large volume of water (and basic materials) present; no additional iodine was volatilized.

Shown together, as in Figure 2.8, the overall iodine distribution would not indicate discernable change from the values at 24 h. However, depicted by itself, the aqueous I_2 inventory shows a steady decline, as seen in Figure 2.11. This effect is due mostly to the gradual oxidation of intermediate species (which are grouped together with I_2) to IO_3^-.
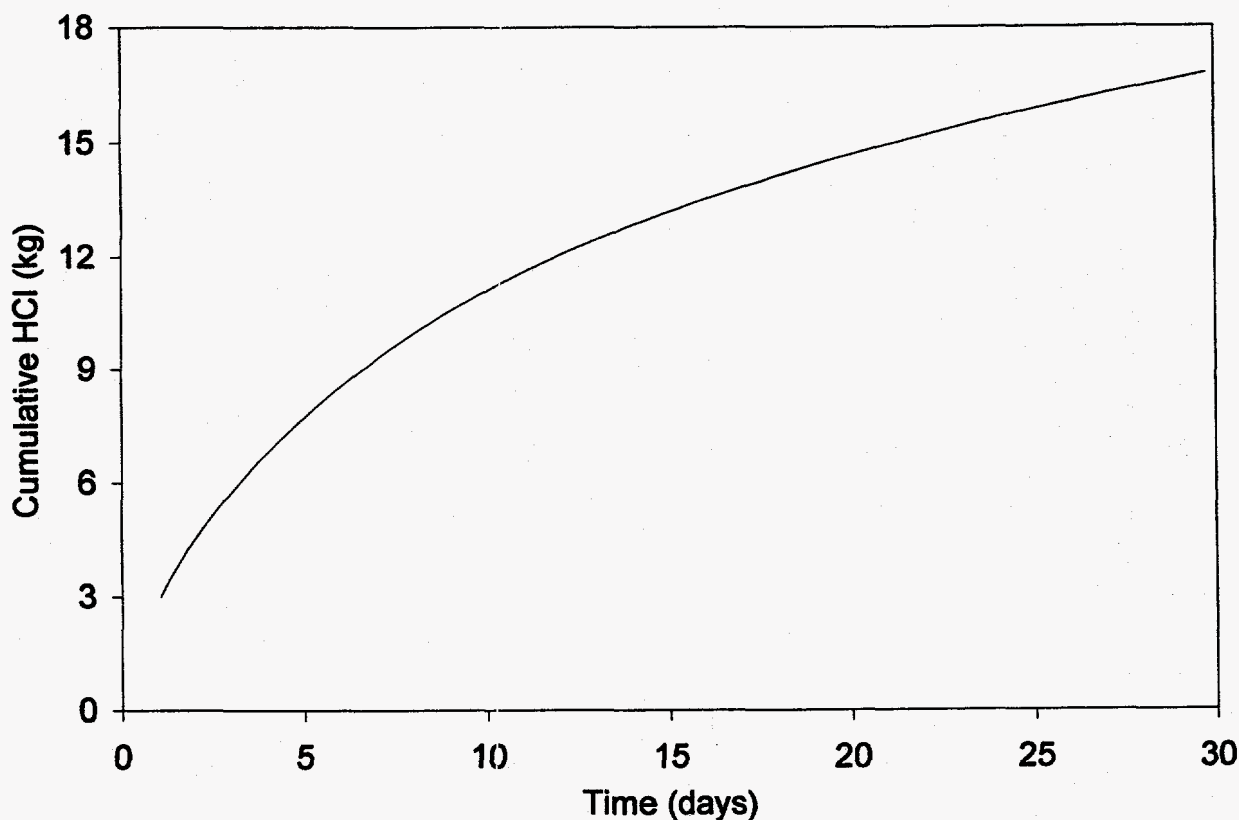

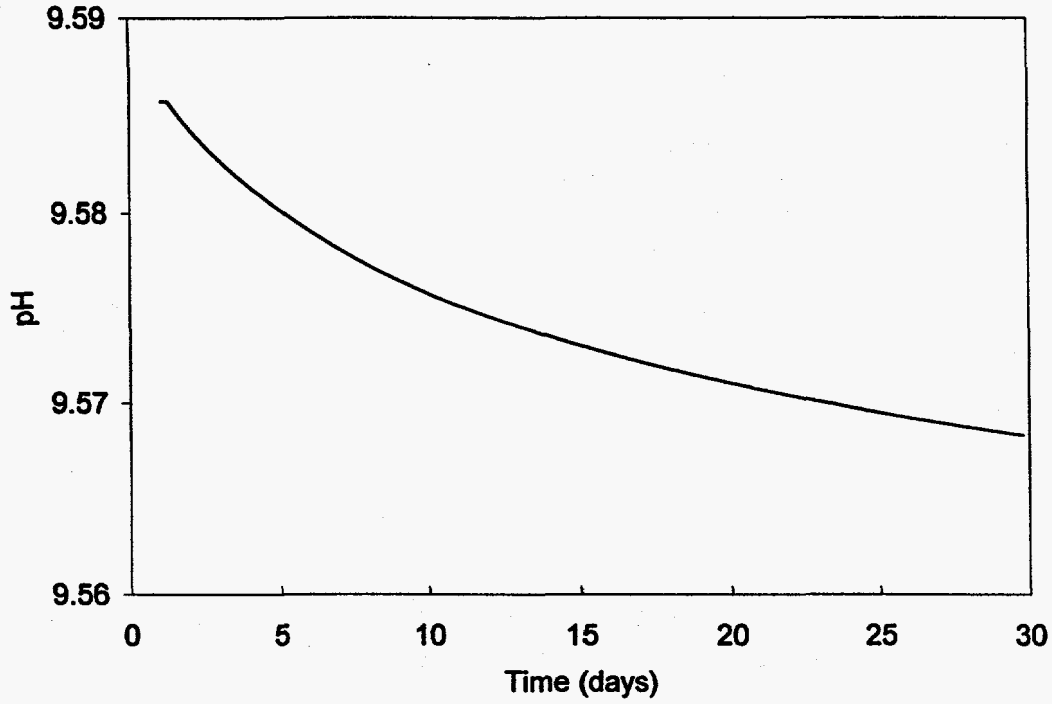**Figure 2.9 Acid production over 30-day period**

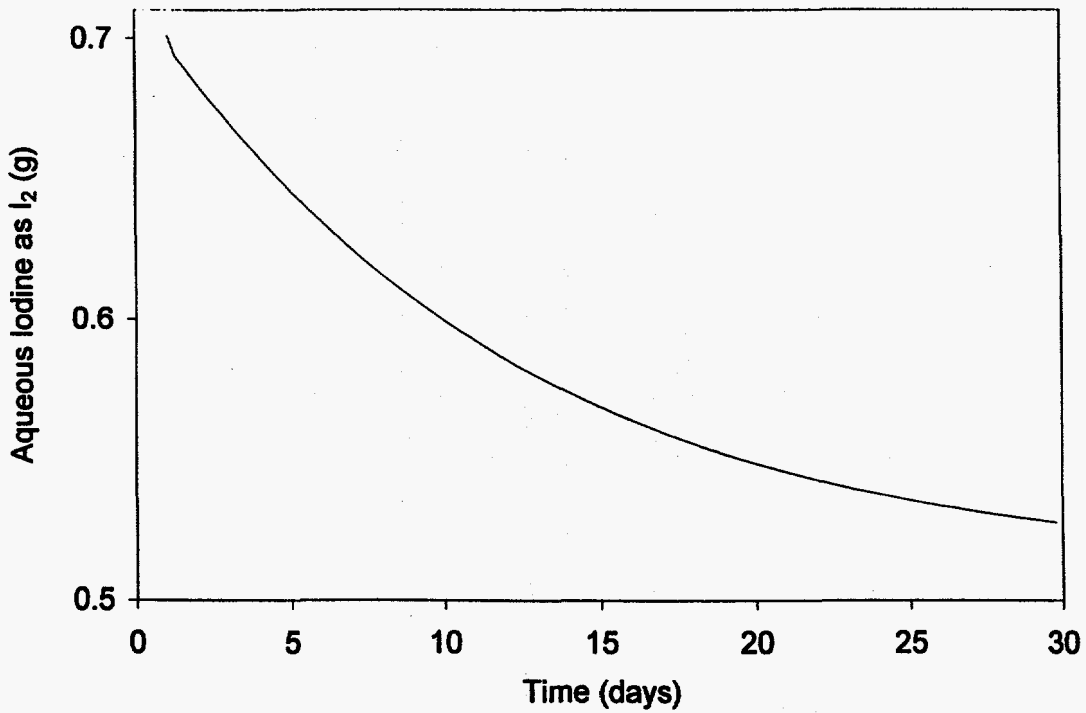**Figure 2.10   Total containment pH over 30-day period**

Figure 2.11   Decline of aqueous volatile inventories

3 Levels of pH in AP-600 Containment Water

The masses of water, acids, and bases in AP-600 containment water were given as follows:

Component	Mass (lb)
H_2O	6.34×10^6
H_3BO_3	1.1×10^5
CsOH	267
HNO_3	211
Na_3PO_4	4567 (10,580 lb of $Na_3PO_4 \cdot 12H_2O$)
HCl	319

With the amount of trisodium phosphate (TSP) indicated above, Westinghouse reported that the pH remained above 7.0 for a 30-day accident duration. In addition, it was noted that the AP-600 basket design was modified to store 11,550 lb of $Na_3PO_4 \cdot 12H_2O$.

To evaluate pH, the masses given by Westinghouse were converted to units suitable for experimental measurement. This resulted in 2.88×10^6 L H_2O, 8.07×10^5 mol H_3BO_3, 8.08×10^2 mol CsOH, 1.52×10^3 mol HNO_3, 1.26×10^4 mol Na_3PO_4, and 3.93×10^3 mol HCl.

The test was based on a solution that contained 500 mL H_2O, 8.685 g H_3BO_3, 0.8357 g $Na_3PO_4 \cdot 12H_2O$, and 0.150 mL of 1 M NaOH. This solution represents a ~0.5-L scale of the Westinghouse masses for water, boric acid, and trisodium phosphate dodecahydrate. In the test, sodium hydroxide was substituted for cesium hydroxide on a mole-for-mole basis. In this solution, the difference between the relatively small amount of sodium hydroxide and the same molar amount of cesium hydroxide would not be detected. It should also be noted that although the hydroxide was included in the test solutions, it is not likely that CsOH would exist in a pure form in containment during an accident sequence. This was discussed in Section 2.3.1 of NUREG/CR-5950.[2] The pH of this solution[*] was measured as 6.87. Following the pH measurement, an additional increment of 0.0754 g $Na_3PO_4 \cdot 12H_2O$ was added to make the solution correspond to the difference between 10,580 lb and 11,550 lb of $Na_3PO_4 \cdot 12H_2O$. The latter amount was given by Westinghouse as the total TSP to be stored in two baskets. After the addition of this increment of TSP, a pH of 6.91 was measured.

After this pH measurement, the solution was titrated with 0.1 M nitric acid (HNO_3). The results of the titration are shown in Table 3.1 and Figure 3.1. The combined HCl and HNO_3 from the masses given by Westinghouse would be represented by 0.19 mL of titrant. In the titration, 0.175 mL of titrant gave a pH of 6.84 and 0.264 mL of titrant resulted in a pH of 6.7. Thus, the relative amount of acid in the Westinghouse recipe would yield a pH of ~6.8. In summary, the pH levels measured from the H_2O, boric acid, and TSP were 6.87 and 6.91 for 10,580 and 11,550 lb of $Na_3PO_4 \cdot 12H_2O$, respectively. When the solution was titrated with 0.1 M HNO_3, the pH decreased slightly to ~6.80 with the acid concentration indicated in the Westinghouse recipe.

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[*] All pH measurements were corrected to 25 °C.

Table 3.1   Data for pH levels in AP-600 containment water

8.6847 g H_3BO_3, 0.150 mL 1 M NaOH, and 0.8357 g $Na_3PO_4 \cdot 12H_2O$, pH 6.87; added
another 0.0754 g $Na_3PO_4 \cdot 12H_2O$, pH 6.91; titrated 10 mL solution using 0.1 M HNO_3

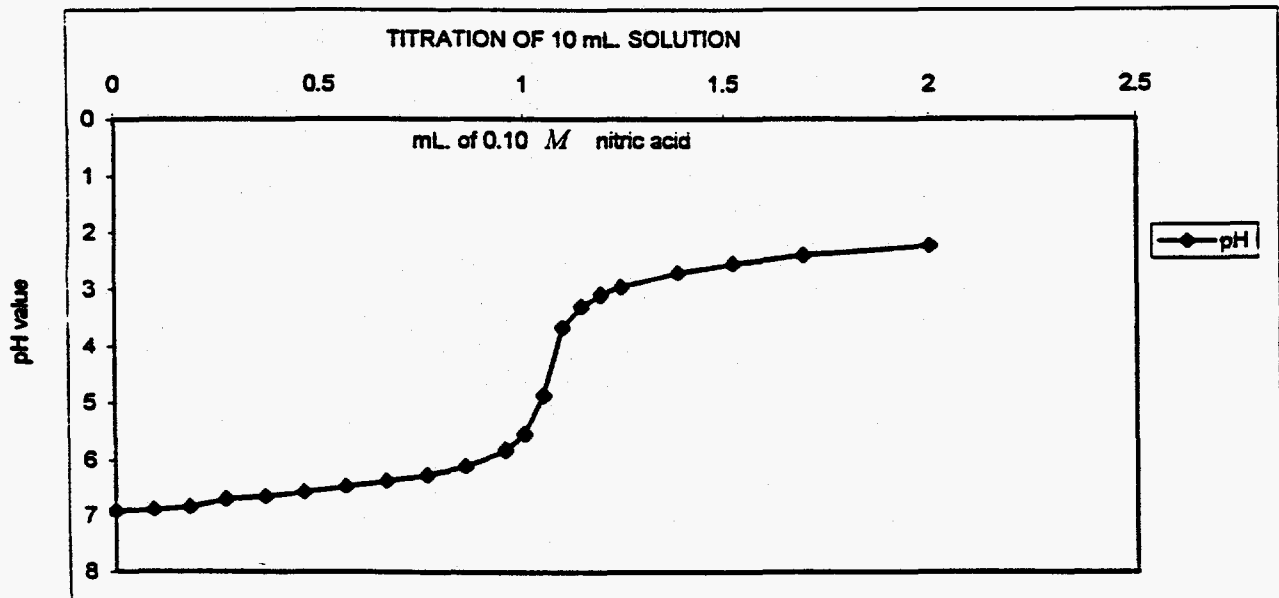| Titrant (mL)	pH
2	2.22
1.696	2.39
1.52	2.54
1.382	2.7
1.239	2.94
1.189	3.08
1.143	3.28
1.096	3.67
1.048	4.86
1.002	5.54
0.953	5.82
0.857	6.1
0.762	6.27
0.661	6.37
0.557	6.48
0.454	6.58
0.359	6.66
0.264	6.7
0.175	6.84
0.088	6.88
0	6.91

**Figure 3.1   pH levels in AP-600 containment water**

4 References

1. Beahm, E. C., C. F. Weber, T. S. Kress, and G. W. Parker, "Iodine Chemical Forms in LWR Severe Accidents," NUREG/CR-5732 (ORNL/TM-11861), Oak Ridge National Laboratory, April 1992.

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5. NUCDECAY—Nuclear Decay Data for Radiation Dosimetry, Calculations for ICRP and MIRD, DLC-172, Radiation Shielding Information Center, Oak Ridge National Laboratory, May 1995. See also K. F. Eckerman et al., "Nuclear Decay Data Files of the Dosimetry Research Group," ORNL/TM-12350, Oak Ridge National Laboratory, 1993, and K. F. Eckerman et al., *Health Phys.* 67(4), 338–345 (1994).

6. Soffer, L. et al., "Accident Source Terms for Light Water Nuclear Power Plants," NUREG-1465, February 1995.
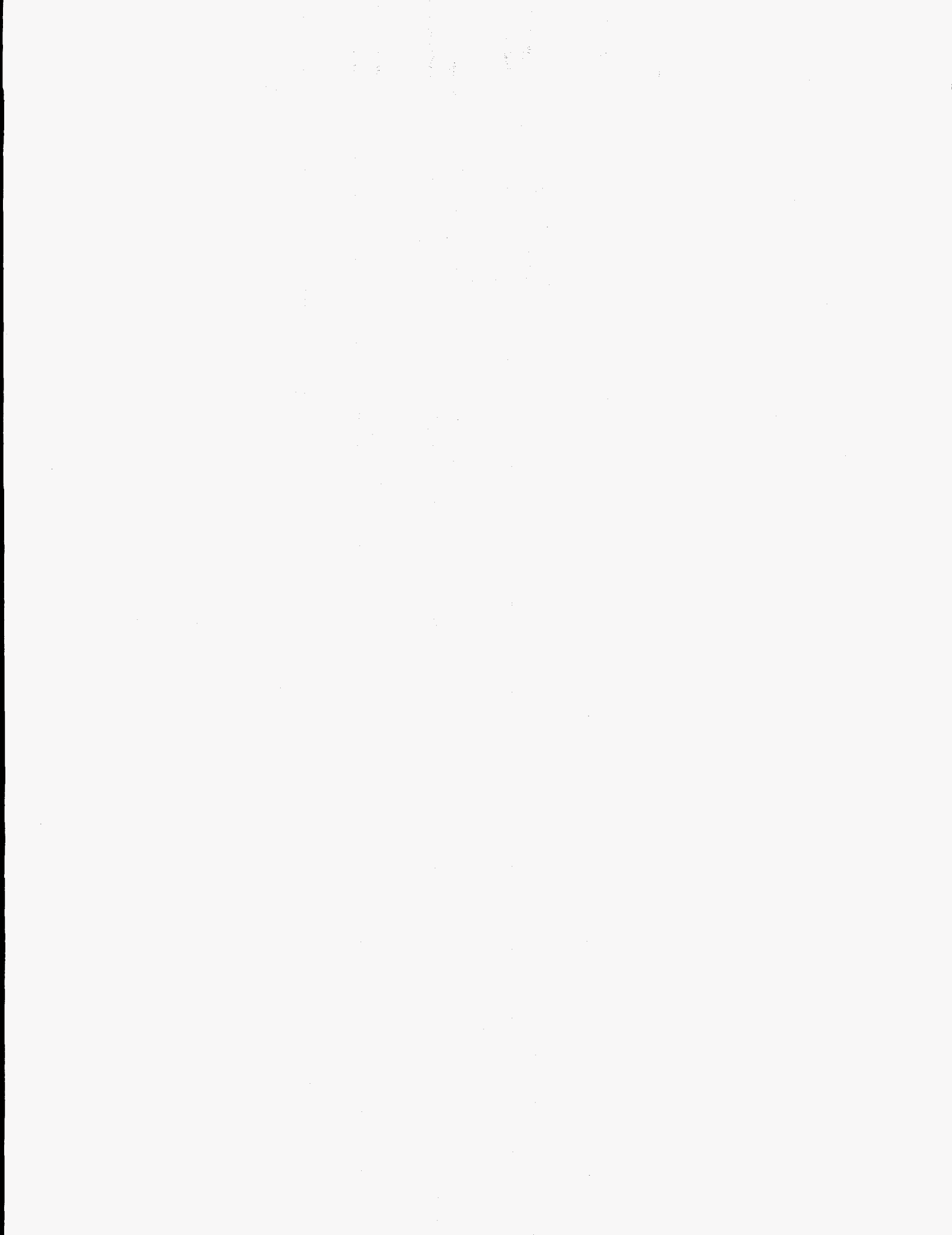
APPENDIX A

RUTHENIUM GROUP RELEASE FRACTION
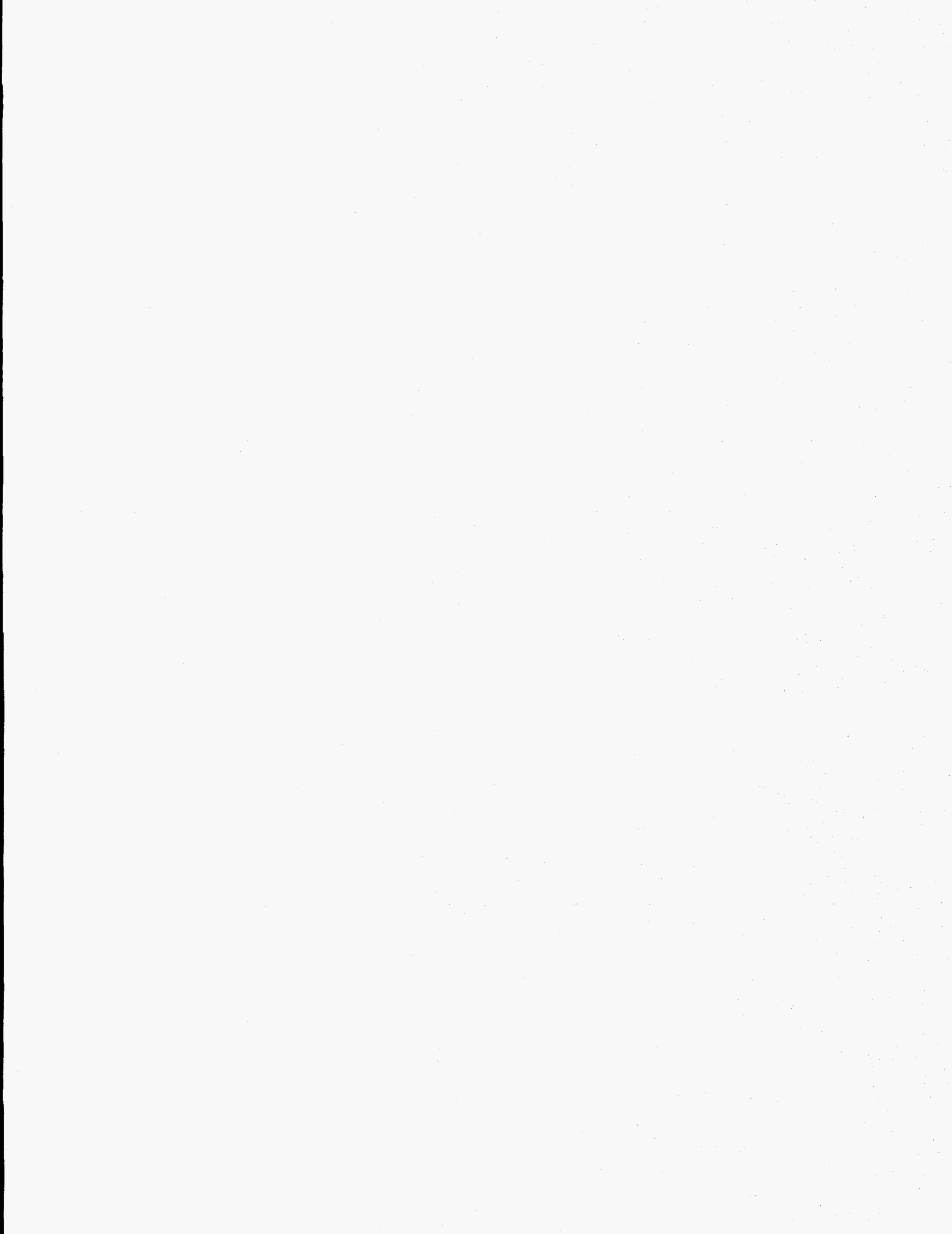
The nuclides comprising the ruthenium group in the MAAP code are listed in Table A.1. The second and third columns give the fractional contribution of each nuclide to group mass and decay energy, respectively. The fourth column lists the in-vessel release fraction applicable to each nuclide, taken from Reference 6. Now, our primary consideration for all groups other than iodine is the contribution to the radiation field in water or air. Hence, the effective in-vessel release fraction for the entire group is

$$F = \sum_i f_i p_i \, ,$$

where f_i = in-vessel release fraction for nuclide i,

          p_i = fraction of group decay energy contributed by nuclide i,

and the summation includes all nuclides in Table A.1. Performing the calculation using values in the table yields F = 0.0009, which is the value appearing in Table 2.3.

Table A.1   Ruthenium group nuclides

| Nuclide | Fraction contributed | | In-vessel relative fraction |
	Mass	Energy	
^{100}Ru	0.014		0.0025
^{101}Ru	0.092		0.0025
^{102}Ru	0.092		0.0025
^{103}Ru	0.004	0.078	0.0025
^{104}Ru	0.061		0.0025
^{105}Ru		0.085	0.0025
^{106}Ru	0.014	0.001	0.0025
^{99}Tc	0.091		0.0025
^{99m}Tc		0.022	0.0025
^{101}Tc		0.003	0.0025
^{104}Tc		0.005	0.0025
^{103}Rh	0.047		0.0025
^{103m}Rh		0.005	0.0025
^{105}Rh		0.021	0.0025
^{106}Rh		0.085	0.0025
^{106m}Rh		0.003	0.0025
^{107}Rh		0.001	0.0025
^{104}Pd	0.029		0.0025
^{105}Pd	0.042		0.0025
^{106}Pd	0.024		0.0025
^{107}Pd	0.023		0.0025
^{108}Pd	0.016		0.0025
^{109}Pd		0.012	0.0025
^{110}Pd	0.005		0.0025
^{95}Nb	0.004	0.134	0.0002
^{97}Nb		0.174	0.0002
^{97m}Nb		0.104	0.0002
^{90}Zr	0.004		0.0002
^{91}Zr	0.071		0.0002
^{92}Zr	0.081		0.0002
^{93}Zr	0.09		0.0002
^{94}Zr	0.093		0.0002
^{95}Zr	0.007	0.14	0.0002
^{96}Zr	0.099		0.0002
^{97}Zr		0.13	0.0002

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10. SUPPLEMENTARY NOTES

J.Y. Lee, NRC Project Manager

11. ABSTRACT *(200 words or less)*

The distribution of iodine in containment during an AP-600 design-basis accident was evaluated using models in the "TRENDS" code. The AP-600 3BE accident sequence calculations showed that a pH greater than 7 was maintained for at least 30 days. Because the pH was maintained at this level, most of the iodine was in the form of iodide; only 3×10^{-3} % was present as aqueous I_2, and only 1×10^{-6} % was present as I_2 in the vapor phase.

12. KEY WORDS/DESCRIPTORS *(List words or phrases that will assist researchers in locating the report.)*

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