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DISSOLUTION OF PLUTONIUM METAL IN SULFAMIC ACID AT ELEVATED TEMPERATURES

L. W. GRAY



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PREPARED FOR THE U.S. DEPARTMENT OF ENERGY UNDER CONTRACT AT(07-2)-1

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DISSOLUTION OF PLUTONIUM METAL IN SULFAMIC ACID AT ELEVATED TEMPERATURES

by

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ABSTRACT

The kinetics of the dissolution of plutonium metal in sulfamic acid at varying temperatures was determined. Hydrogen off-gas rates and plutonium concentrations were determined as functions of time for different dissolving conditions. Nominal 2.2 kg Pu buttons (surface area, 171 cm²) should yield solutions containing 190 to 260 g Pu/L and 3.3M sulfamate ion after a one-hour dissolving time at 70°C. Hydrogen off-gas rates were calculated to vary from a high of 2.7L of gaseous hydrogen per minute (STP) at the beginning of a dissolving cycle to 50 mL of hydrogen gas per minute at the end of a one-hour dissolving time. Sludge formation, which is a problem at ambient temperatures, does not occur at dissolving temperatures of 60 to 80°C. Dissolution curves suggest that the mechanism of dissolution changes at about 50 to 55°C. Below this temperature, a surface coating of plutonium hydride may form; above this temperature, plutonium metal appears to be converted directly to Pu³⁺.

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DISSOLUTION OF PLUTONIUM METAL IN SULFAMIC ACID AT ELEVATED TEMPERATURES

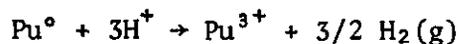
INTRODUCTION

Plutonium metal that does not meet product purity specifications and aged plutonium metal into which the daughter product ^{241}Am has grown must be recycled through a recovery and purification process. At the Savannah River Plant (SRP), the initial step in this recycle is the conversion of the metal to an aqueous solution. Since about 1962, sulfamic acid has been the accepted dissolvent for plutonium metal in the SRP process.¹

Previous studies¹ have shown that the rate of dissolution at ambient temperatures is dependent on the hydrogen ion concentration and the surface area of the metal. When 1.67M sulfamic acid is the dissolvent, plutonium concentrations as high as 134 g/L can be obtained.^{2,3} In normal production practice, however, concentration of 60 ± 10 g Pu/L are normally obtained in one-hour.¹

SUMMARY AND CONCLUSIONS

Plutonium metal dissolves readily in sulfamic acid ($\text{NH}_2\text{SO}_3\text{H}$) solutions at temperatures from ambient to about 80°C according to the reaction



In the lower temperature range (ambient to about 50°C), dissolution appears to proceed through a plutonium hydride intermediate. In the temperature range of 50 to $\sim 80^\circ\text{C}$, the intermediate apparently does not form. As a result, metal dissolved at temperatures between 60 and 80°C does not yield a sludge. The sludge normally contains about 0.1 to 3% of the metal when dissolved at ambient temperatures. Dissolution of plutonium metal at temperatures greater than about 80°C was not attempted because the evaporated water from the in-plant process could create problems within the off-gas and vacuum-transfer systems.

The results reported here indicate the optimum dissolution temperature is in the range of 65 to 80°C . Within this range, plutonium concentrations of 180 to 250 g Pu/L can be obtained. This requires dissolving times of about one hour and a sulfamic

acid concentration of 3.34M. By coupling sulfamic acid precipitation, sulfamic acid concentration, and higher temperature dissolution, the sulfate-to-plutonium mole ratio in Purex feed can be reduced from the nominal 6.7-to-one as found with low temperature dissolution to a mole ratio of about one-to-one.

Reducing the sulfate-to-plutonium ratio has several advantages:

1. Lower waste volume to be stored.
2. Less corrosion of the evaporators.
3. Fewer complications in the recovery of ^{241}Am , when ^{241}Am is to be recovered from the waste streams.

Increasing the dissolution temperature has the following advantages:

1. Faster throughput rate.
2. Less sludge generated; hence, the safety hazard of dissolving plutonium metal is removed.
3. A lower sulfamate-to-plutonium ratio in the dissolver solution; hence, less evaporator corrosion and reduced waste volume.

For calculational purposes, a typical dissolving cycle was assumed to be:

1. Charge a 2.250 kg plutonium button (171.2 cm² surface area) to 3.0 L of sulfamic acid at 70°C.
2. Allow dissolution to proceed for 60.0 min.
3. Displace 2.0 L of this solution with 2.0 L of fresh sulfamic acid.
4. Add another plutonium button.
5. Repeat Steps 2 and 3 until the plutonium metal inventory decreases to 1750 g Pu, then repeat Step 4.
6. Repeat Step 5 as long as necessary.

Dissolving rates obtained in these experiments were used to calculate the hydrogen off-gas rates. These calculated off-gas rates varied from as high as about 2.7 L/min of hydrogen at the beginning of the dissolving time to less than 50 mL/min of hydrogen near the end of a one-hour dissolving time.

EXPERIMENTAL PROCEDURE

The volume of hydrogen gas generated as a function of time was measured for all experiments by measuring the volume of water displaced from a gas-tight reservoir (Figure 1). The volume of water displaced was measured with a graduated cylinder. Reaction rates for plutonium were determined on pieces of plutonium metal cut from rolled sheet. The surface area of these pieces was calculated from dimensions measured with calipers. The sulfamic acid solution was equilibrated to the temperature of a waterbath at the desired reaction temperature before introducing the metal.

CALCULATION OF BUTTON SURFACE AREA

SRP plutonium buttons are produced in a ceramic-lined bomb-reduction furnace. A typical button is 0.75-in. thick, weighs about 2250 g, and has a density of 19.3 g/cm^3 . The equations for the surface area and volume of SRP-produced buttons were developed in an earlier paper.¹ Both the surface area and the volume of a typical button as a function of plutonium remaining undissolved are shown in Figure 2.

DISSOLVING RATES OF PLUTONIUM METAL

Hydrogen generation with time was determined for a series of plutonium metal pieces at varying temperatures. Typical curves for these dissolutions are shown in Figure 3. The rate of evolution was determined from the tangent of the early portion of the curve for evolved hydrogen (Figure 4). This value was divided by the initial surface area of the plutonium metal piece being dissolved to determine the rate of hydrogen evolved in terms of surface area $[\text{mL H}_2(\text{g})/(\text{min} \cdot \text{cm}^2)]$.

Figure 4 shows a sharp change in slope of the dissolution curve at about 50°C . If the dissolving mechanism were the same throughout the temperature region, one would expect a constant slope. Also, the formation of sludge ceases at the same temperature. This can, perhaps, be interpreted as follows. Below about 50°C , the dissolution proceeds through a plutonium hydride intermediate, PuH_2 ; above about 50°C , this intermediate does not form. This intermediate would explain why the dry sludge is pyrophoric as well as shock sensitive. It would also explain the release of a gas and the report when the dry sludge suddenly ignites, and why only plutonium dioxide is found in passivated sludge.

The sulfamic acid concentration fed to the dissolver is not altered during the dissolution process. However, as the plutonium

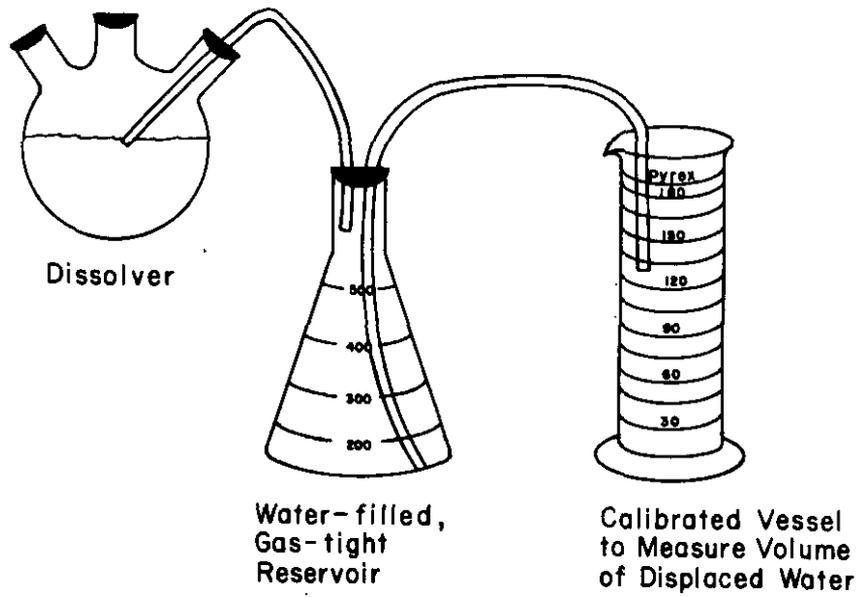


FIGURE 1. Experimental Apparatus

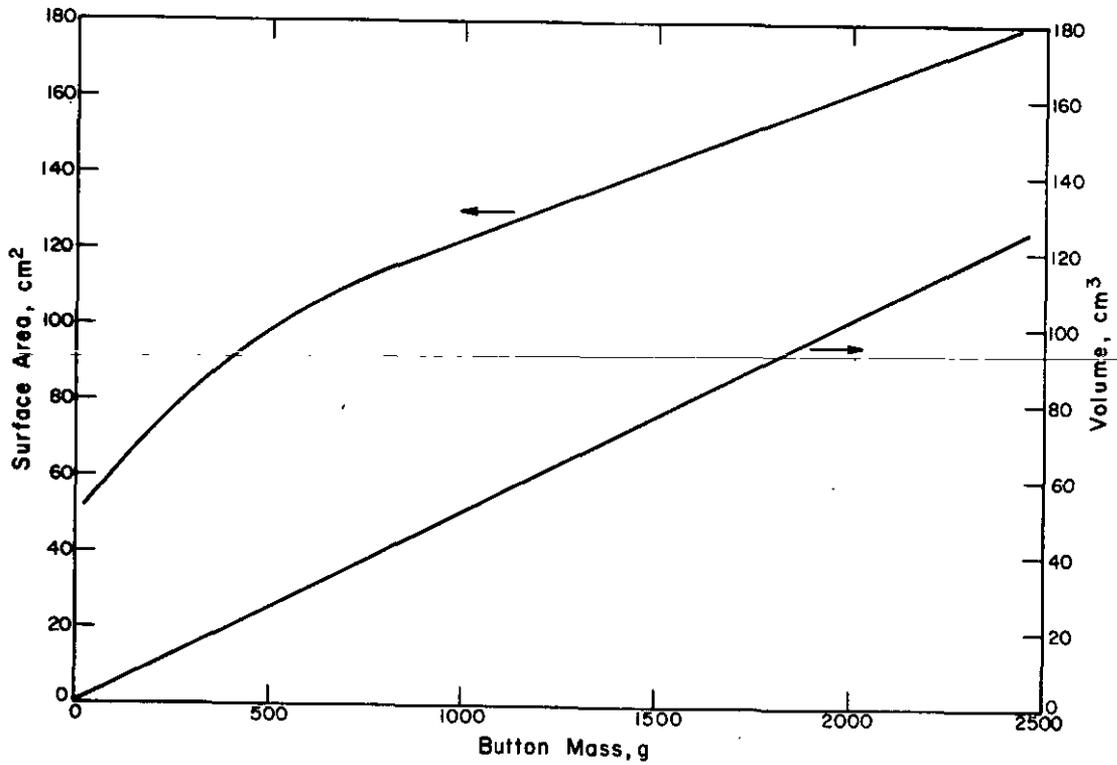


FIGURE 2. Calculated Surface Area and Volume of a Typical Plutonium Button During Dissolution

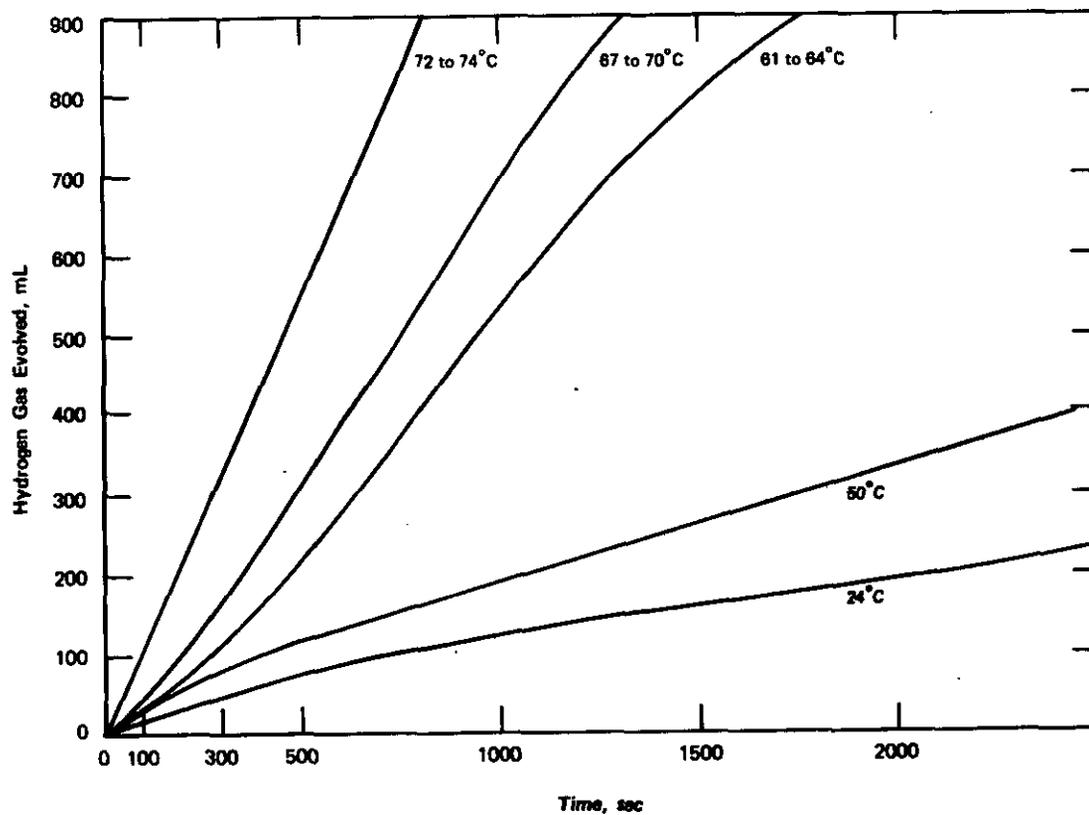


FIGURE 3. Typical Hydrogen Evolution Curves During Plutonium Dissolutions

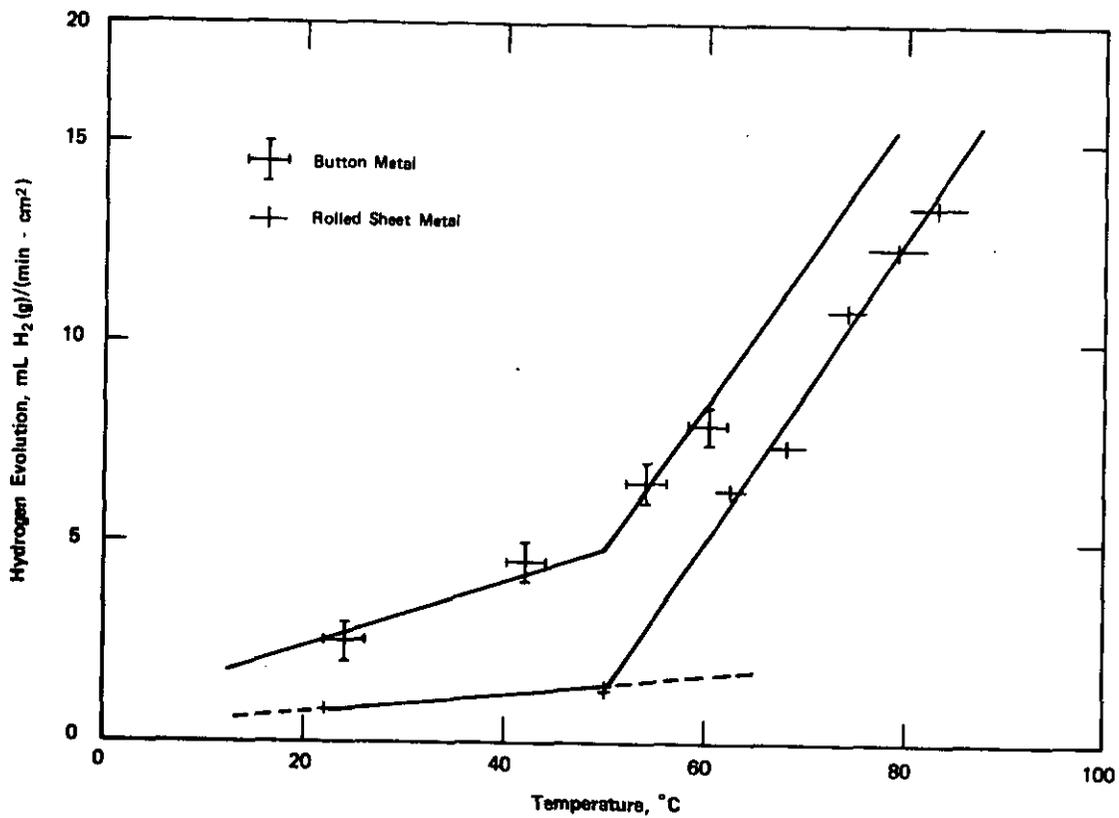


FIGURE 4. Rate of Hydrogen Evolution versus Temperature During Dissolution of Plutonium in 1.67M Sulfamic Acid

metal dissolves, the sulfamic acid concentration in the dissolver is reduced by the reaction. For this reason, the hydrogen evolution rate in mL H₂ (g)/(sec · cm²) of surface area was determined as a function of sulfamic acid concentration at a fixed temperature (Figures 5 and 6). The temperature for this dissolution (70°C) was chosen on the following bases:

1. No sludge is formed at this temperature.
2. Reaction rates are rapid.
3. Vapor pressure of the aqueous solution is still relatively low.

Note (Figure 6) the change in slope of the hydrogen evolution rate curve which occurs at about 1.6M sulfamic acid. As there is no further increase in dissolution rate with increasing sulfamic acid concentration, this change is interpreted as a change in the rate-controlling phenomenon. Below about 1.6M sulfamic acid, the rate-controlling phenomenon is the diffusion of hydrogen ions to the surface of the metal. Above about 1.7M sulfamic acid, the rate-controlling phenomenon is the diffusion of gas bubbles away from the surface of the metal.

A nominal 2250g button is dissolved in a minimum of three separate batches to avoid nuclear safety (criticality) problems. Each of these three batch operations are discussed individually and are summarized on Table 1. However, the calculations which follow were made using the rates measured for plutonium sheet metal and are considered more conservative, i.e., they are less than those for plutonium buttons. The effect of increasing the initial sulfamic acid concentration from 1.67M to 3.34M is also discussed.

Case 1: Dissolution of One Plutonium Button in 1.67M Sulfamic Acid (Batch 1)

The initial maximum rate of dissolution and of hydrogen gas evolution (at startup) would be predicted to occur if the initial button were charged to 1.67M sulfamic acid at 70°C. Calculations show that this maximum hydrogen evolution rate would be about 1.5 L of hydrogen per minute. After a 0.25-hr dissolving time, the plutonium concentration would have increased to about 44 g/L (Figure 1), and the hydrogen evolution rate would have decreased to about 1.0 L/min (Figure 7). This is about the same plutonium concentration obtained after about the first 0.75 hour of dissolving of the initial batch at ambient temperature.

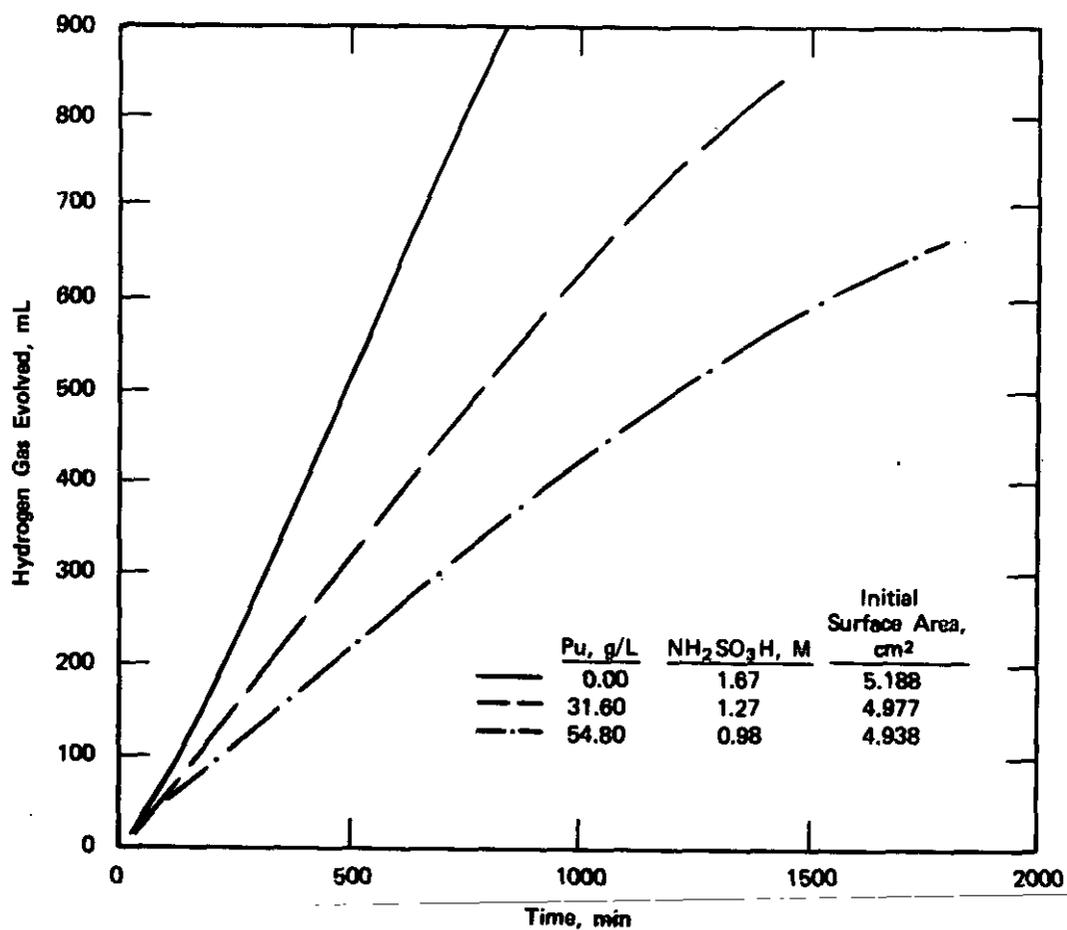


FIGURE 5. Hydrogen Evolution Curves at Varying Plutonium and Sulfamic Acid Concentrations

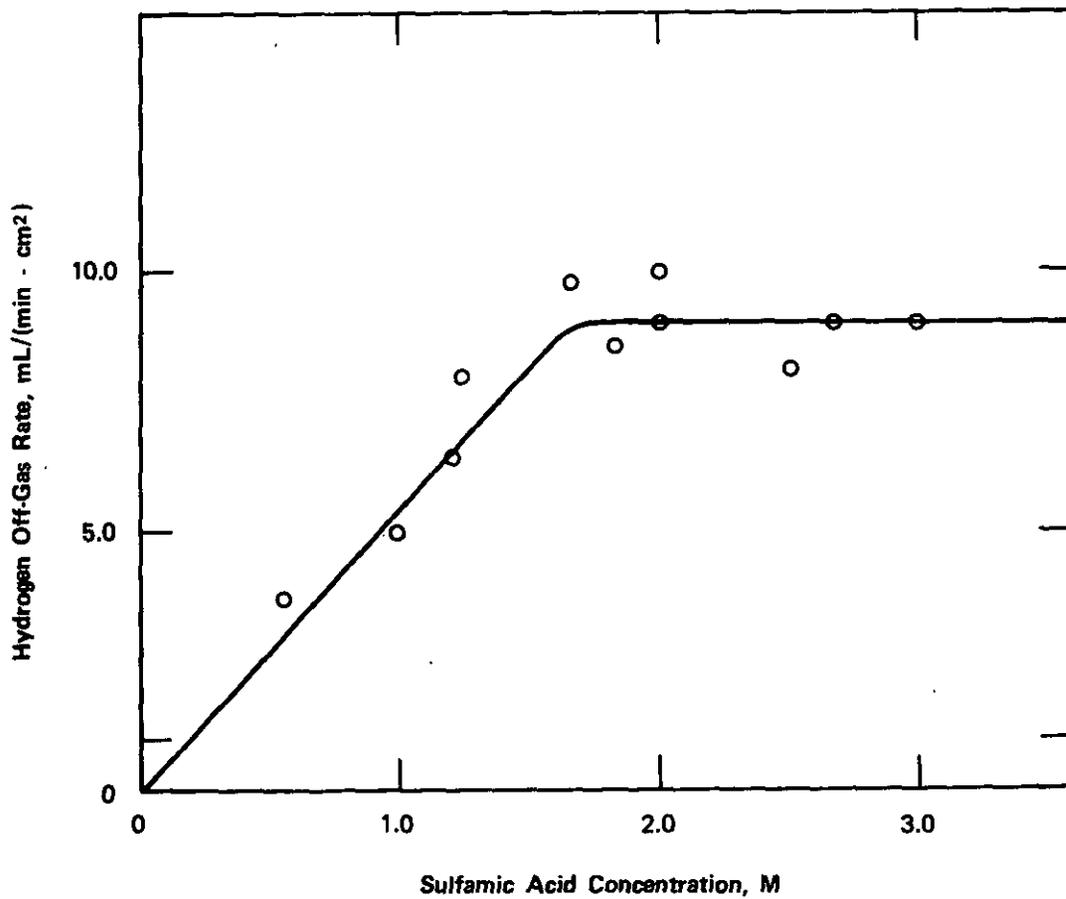


FIGURE 6. Dissolution Rate of Plutonium at 70°C versus Sulfamic Acid Concentration

TABLE 1

Dissolution of Plutonium Metal in Sulfamic Acid at 70°C

Case No.	<i>Initial Conditions</i>		<i>Conditions at 60 min</i>					
	<i>Surface Area, cm²</i>	<i>Pu metal, g</i>	<i>Pu in sol'n, g/L</i>	<i>Sulfamic acid, M</i>	<i>Pu in sol'n, g/L</i>	<i>Sulfamic acid, M</i>	<i>SA/Pu Ratio^a</i>	<i>Dissolution Time, min</i>
1	171.2	2246.15	0.00	1.67	105.06	0.352	3.80	60
2	158.7	1930.98	35.02	1.23	110.34	0.286	3.62	60
3	149.9	1705.03	36.78	1.21	108.76	0.305	3.67	60
4	312.3	3735.24	36.25	1.21	128.28	0.060	3.11	60
5	171.2	2246.15	0.00	3.34	187.28	0.990	4.26	60
					226.66	0.496	3.52	90
6	144.4	1566.18	75.55	2.39	210.76	0.696	3.79	60
					238.00	0.350	3.36	90
7	296.4	3325.15	79.33	2.34	253.61	0.151	3.15	60

a. The average sulfamic acid/plutonium metal ratio (SA/Pu) for dissolving plutonium in 1.67M sulfamic acid at ambient temperature is 6.65.

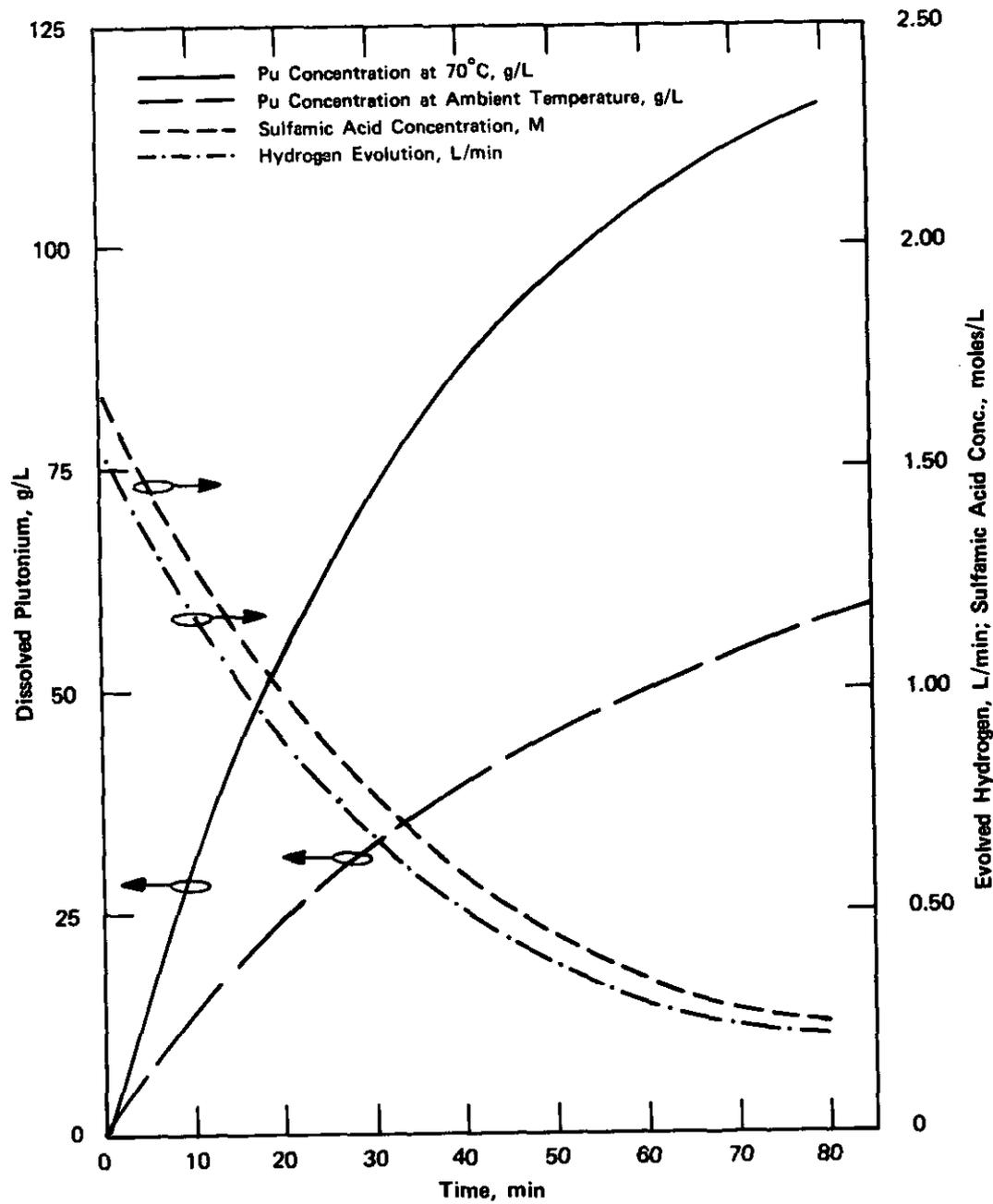


FIGURE 7. Batch Dissolution of Plutonium in 1.67M Sulfamic Acid at 70°C (Case 1)

After a 0.5-hr dissolving time, the plutonium concentration was calculated to have increased to about 73 g Pu/L and the hydrogen evolution rate decreased to about 0.7 L/min. At the end of an hour, the plutonium concentration increased to about 105 g Pu/L and hydrogen evolution decreased to about 0.3 L/min.

Higher dissolving rates for plutonium metal could be obtained if a higher dissolving temperature were used. However, the vapor pressure of water over the dissolving medium would also increase. To maintain the evaporation rate of solution within the process off-gas limits, the temperature should remain below about 80°C.

For convenient comparison, the dissolution curve for a plutonium button in 1.67M sulfamic acid at ambient temperature is also included in Figure 7.

Case 2: Continued Dissolution of One Plutonium Button (Batch 2)

The second dissolving batch is calculated to begin with a residual plutonium concentration of about 35 g Pu/L, a residual sulfamic acid concentration of 1.52M, a button surface area of 158.7 cm², and a hydrogen off-gas rate of about 1.0 L/min. After a 0.25-hr dissolving time, the hydrogen evolution rate is calculated to have decreased to about 0.7 L/min (Figure 8). The dissolved plutonium concentration is calculated to have increased to about 65 g Pu/L (Figure 8); this is about the same plutonium concentration as that obtained after a one-hour dissolving time at ambient temperature. Again for convenience in comparison, the dissolution curve for the second dissolving batch at ambient temperature is included in Figure 8. At the end of one hour of dissolution, the plutonium concentration is calculated to have increased to about 110 g Pu/L and the hydrogen evolution has decreased to about 0.23 L/min.

Case 3: Continued Dissolution of One Plutonium Button (Batch 3)

Calculations show that a third dissolving batch will be necessary to reduce the plutonium inventory in the dissolver to less than 1700 g Pu. This third batch would proceed much as the second batch. It would begin with a residual plutonium concentration of about 36.8 g Pu/L, a residual sulfamic acid concentration of 1.21M, a button surface area of 149.9 cm², and a hydrogen off-gas rate of about 0.95 L/min. After one-hour of dissolution, the plutonium concentration is calculated to have increased to about 108 g Pu/L. These data are shown in Figure 9.

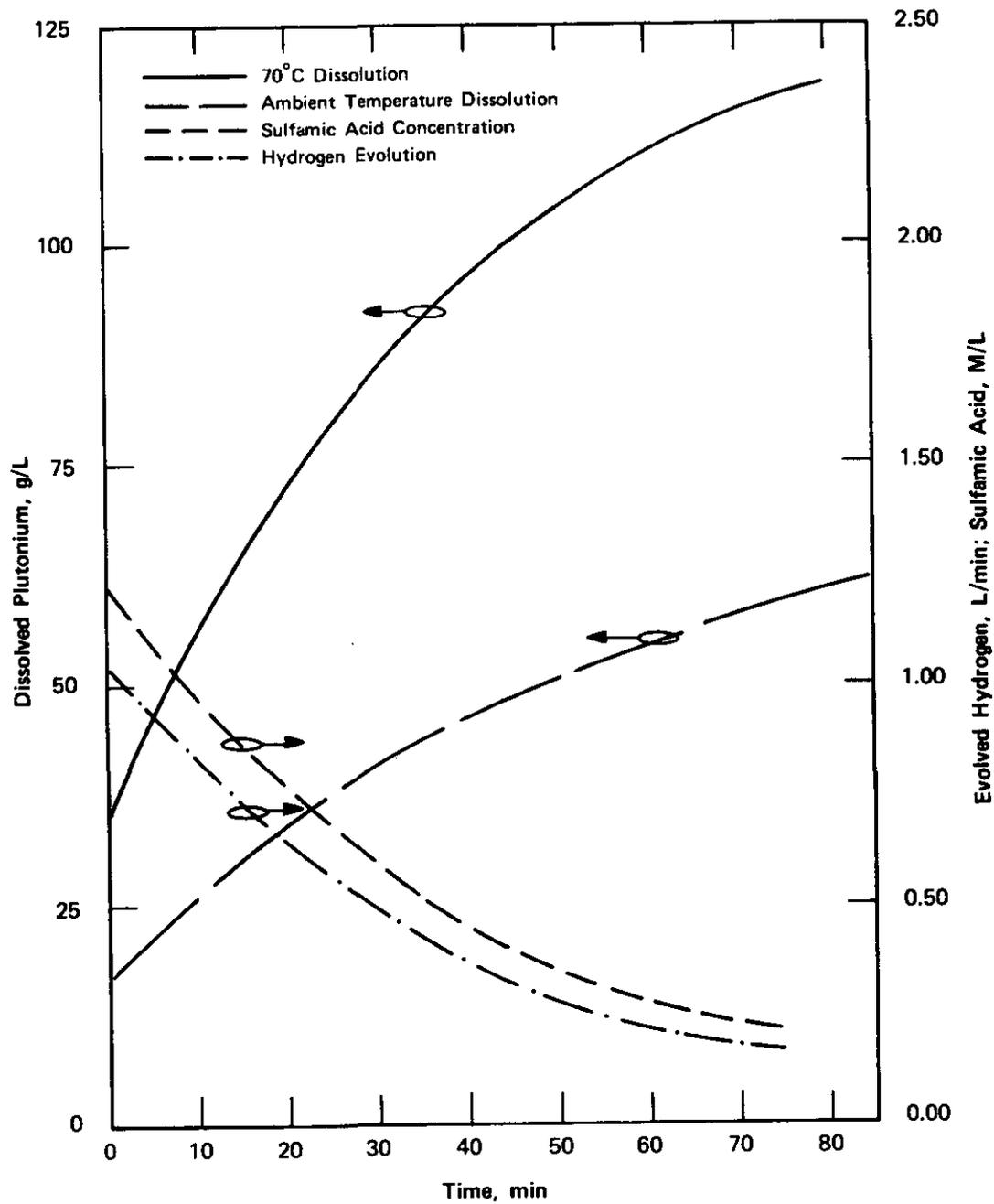


FIGURE 8. Batch Dissolution of Plutonium in 1.67M Sulfamic Acid at 70°C (Case 2)

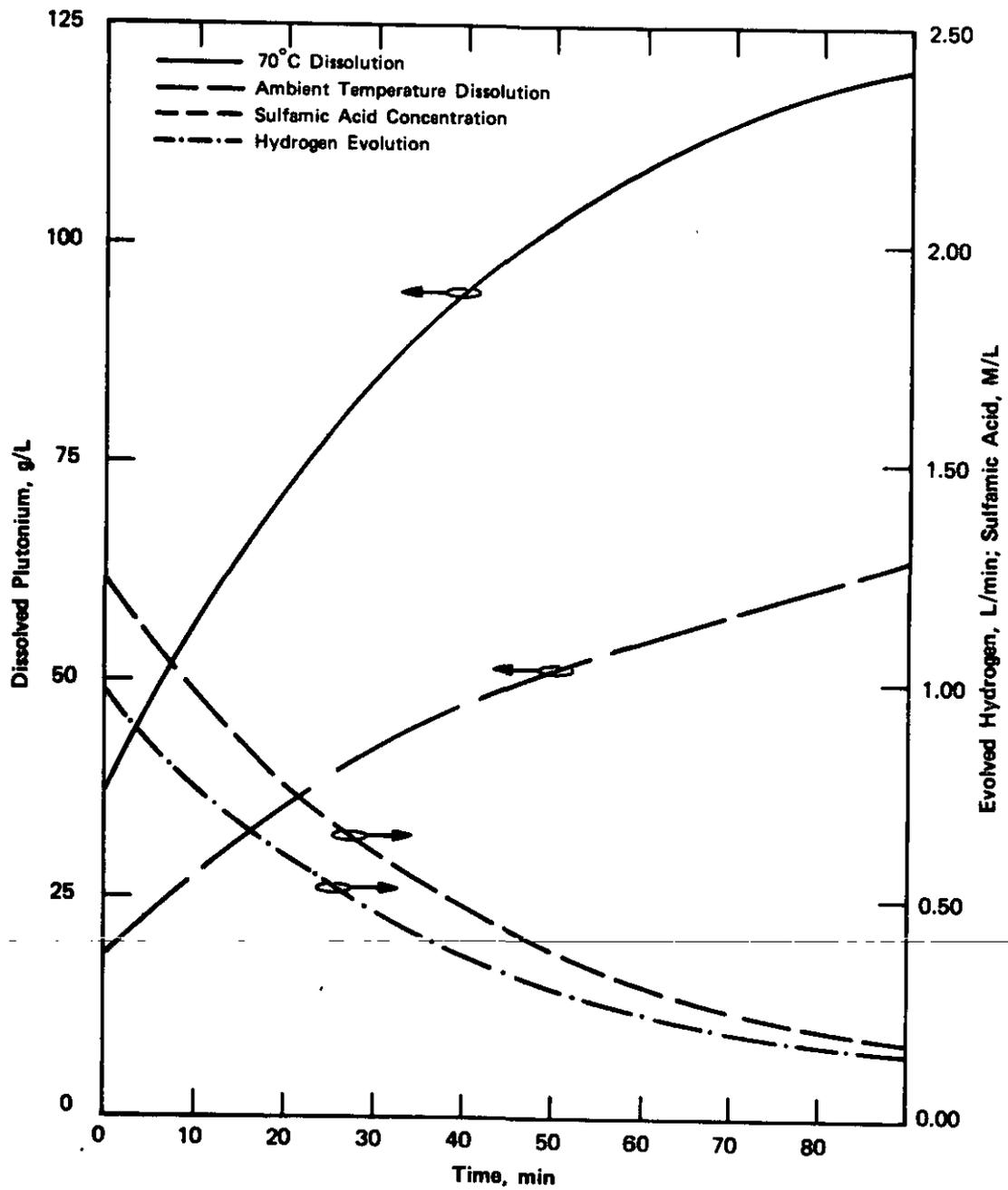


FIGURE 9. Batch Dissolution of Plutonium in 1.67M Sulfamic Acid at 70°C (Case 3)

Case 4: Continued Batch Dissolution of Plutonium Buttons (Batch 4)

After the third dissolving batch, the plutonium inventory of the dissolver has been sufficiently reduced to allow a second button to be added to the dissolver. This button increases the surface area available to the solution for dissolving from 141.1 cm² to 312.3 cm². For calculational purposes, this batch was assumed to begin with a residual plutonium concentration of about 36.25 g Pu/L, a residual sulfamic acid concentration of 1.215M, a button surface area of 312.3 cm², and a hydrogen off-gas rate of about 2.05 L/min. After a 0.25-hr dissolving time, the plutonium concentration is calculated to have increased to about 87.9 g Pu/L and the hydrogen evolution rate would have decreased to about 0.99 L/min (Figure 10). At the end of one hour of dissolution, the plutonium concentration would be about 128 g/L, and the hydrogen evolution rate reduced to 0.08 L/min. This plutonium concentration compares to only about 60 g Pu/L for the same conditions at ambient temperature (Reference 1 and Figure 10).

Case 5: Dissolution of One Plutonium Button in 3.34M Sulfamic Acid (Batch 1)

A sulfamic acid concentration of 1.67M was chosen for the original production dissolvers because this is a near-saturated solution at ambient temperatures. However, for elevated temperature operations, higher sulfamic acid concentrations can be used. At 70°C, a 3.34M sulfamic acid concentration is near saturation. Calculations for the following cases, therefore, assume a sulfamic acid concentration of 3.34M and a dissolution temperature of 70°C.

Calculations show that the maximum hydrogen evolution rate would be about 1.5 L/min. In contrast to the 1.67M sulfamic acid dissolution, the hydrogen evolution rate after 0.25 hr will be about 1.5 L/min, instead of having dropped to about 1.0 L/min. After 0.5 hr, the hydrogen evolution rate will drop only 0.1 L/min (to about 1.4 L/min). At the end of a 1.0-hr dissolving time, the plutonium concentration will increase to almost 190 g Pu/L, instead of only ~105 g Pu/L for the 1.67M sulfamic acid dissolution at 70°C (Figure 11). For dissolving at ambient temperature in 1.67M sulfamic acid, the plutonium concentration on this first batch dissolution reaches only about 45 to 50 g Pu/L. The overall dissolution rate will, therefore, increase by a factor of about 4.

For convenience of comparison, the calculated plutonium concentration with time for 1.67M sulfamic acid both at ambient temperature and at 70°C is included in Figure 11.

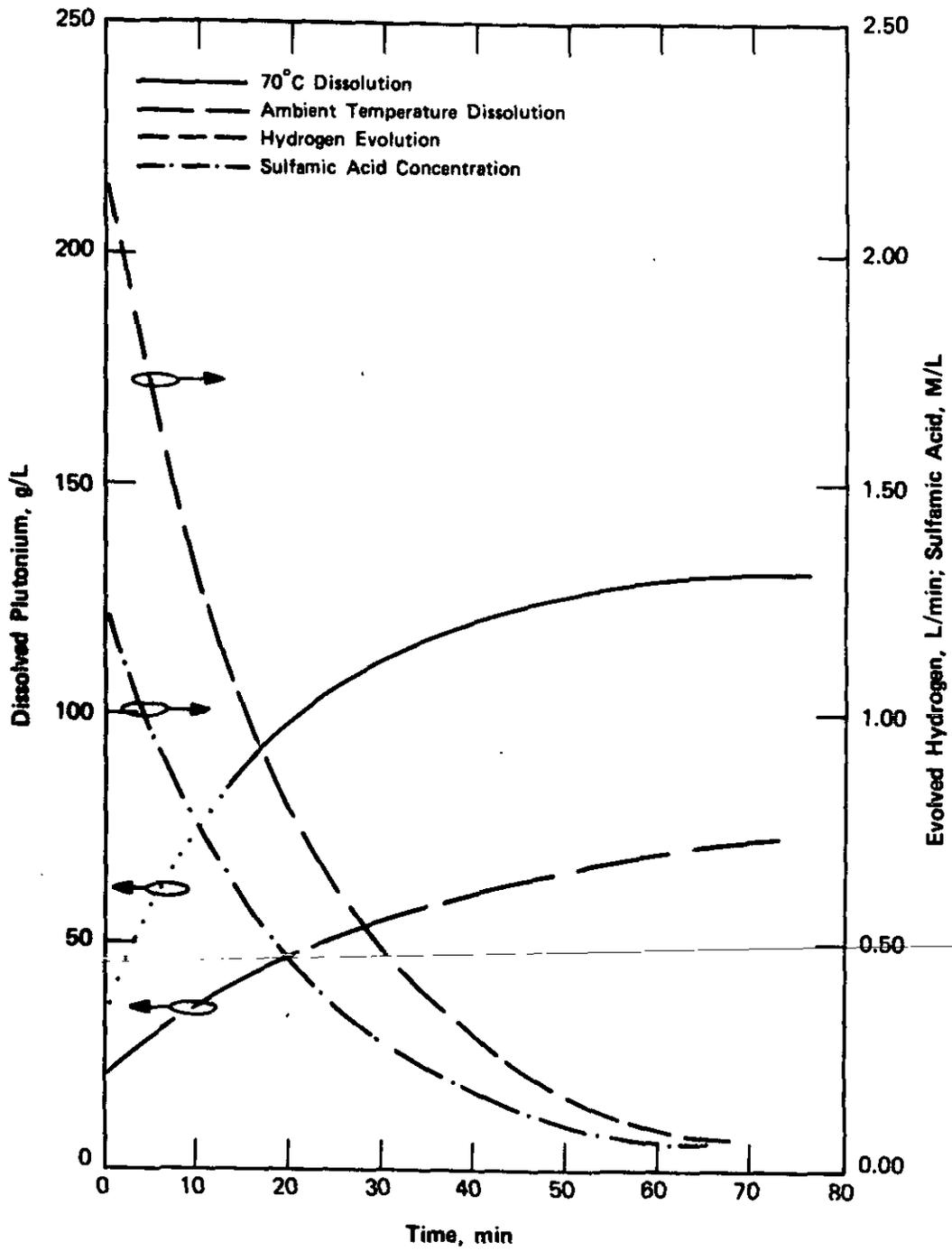


FIGURE 10. Batch Dissolution of Plutonium in 1.67M Sulfamic Acid (Case 4)

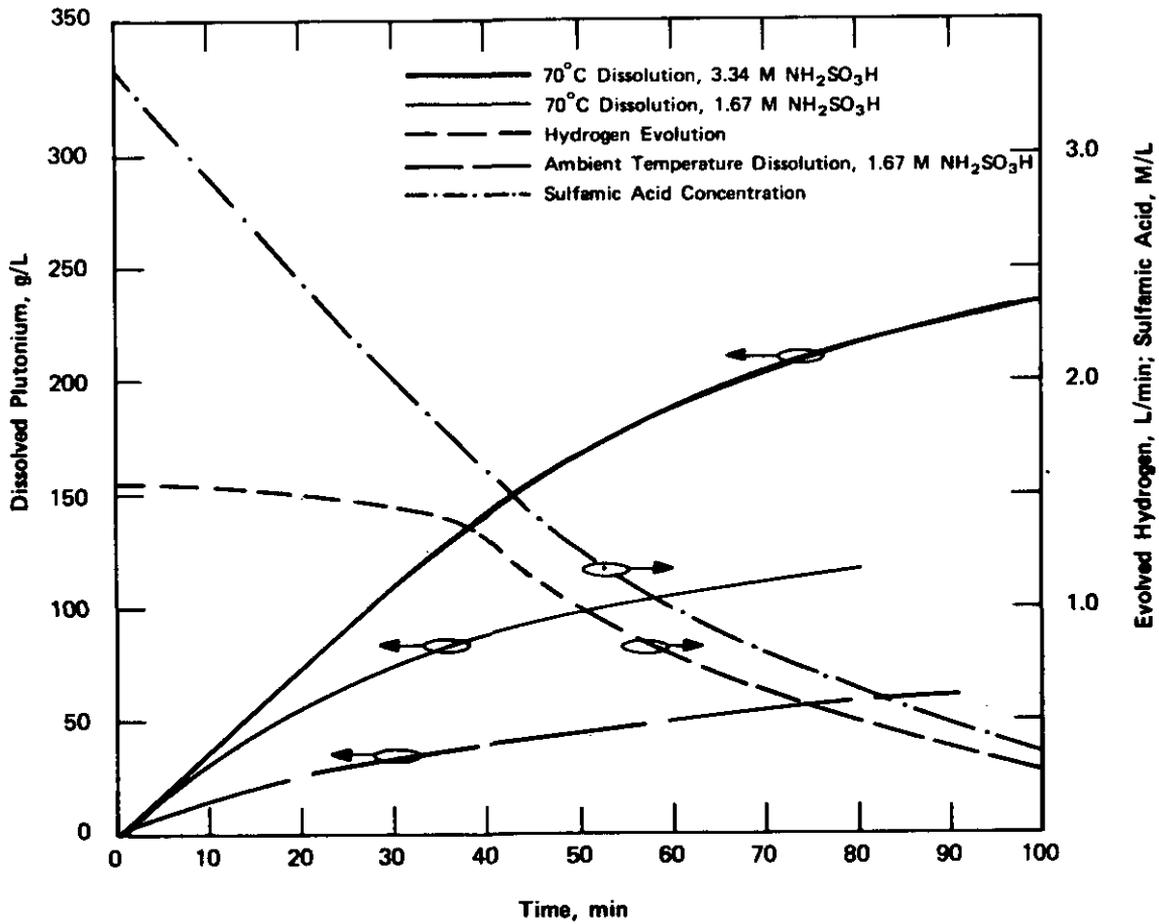


FIGURE 11. Batch Dissolution of Plutonium in 3.34M Sulfamic Acid (Case 5)

For overall efficiency of downstream processing, a dissolving time of 90 minutes should be considered for the initial dissolving sequences. The 90-minute cycle time will increase the dissolved concentration from about 190 g Pu/L to about 225 g Pu/L. The sulfamate-to-plutonium mole ratio will decrease from 4.3 to 3.5. (In contrast, the average sulfamate-to-plutonium mole ratio for 1.67M sulfamic acid/ambient temperature dissolution is 6.6.¹)

Case 6: The Second Dissolving Batch with 3.34M Sulfamic Acid

The second dissolving batch is assumed to begin after a 90-minute dissolving time. The dissolution is calculated to begin with a plutonium concentration of about 75.6 g Pu/L, a sulfamic acid concentration of 2.39M/L, a plutonium button surface area of 144.4 cm², and a hydrogen off-gas rate of about 1.30L/min. After 0.25 hr, the hydrogen evolution rate is calculated to have decreased to about 1.25 L/min (Figure 12), and the dissolved plutonium concentration is calculated to have increased to about 121 g Pu/L. After a 1.0-hr dissolving time, the plutonium concentration is calculated to have increased to about 211 g Pu/L, and the hydrogen evolution has decreased to about 0.50 L/min. In contrast, the plutonium concentration of the 1.67M sulfamic acid/70°C dissolution is calculated to have reached only about 110 g Pu/L. The 1.67M sulfamic acid/ambient temperature dissolution reaches only about 60 g Pu/L in the same time interval. In addition to an overall increase in dissolving rate of about 3.5, the sulfamate-to-plutonium mole ratio has also decreased from 6.65 to about 3.8.

For convenience of comparison, the calculated plutonium concentrations with time for 1.67M sulfamic acid both at ambient temperature and 70°C are included in Figure 12.

For overall efficiency of downstream processing, a dissolving time of 90 minutes should also be considered for the second dissolving cycle. Use of the 90-minute time interval would increase the plutonium concentration from about 211 g Pu/L to about 238 g Pu/L. This would further decrease the sulfamate-to-plutonium mole ratio from 3.8 to 3.4.

Case 7: The Third Dissolving Batch with 3.34M Sulfamic Acid

After the second batch dissolution in 3.34M sulfamic acid at 70°C, the plutonium inventory of the dissolver has been sufficiently reduced to allow a second button to be added to the dissolver. This increases the surface area available to the

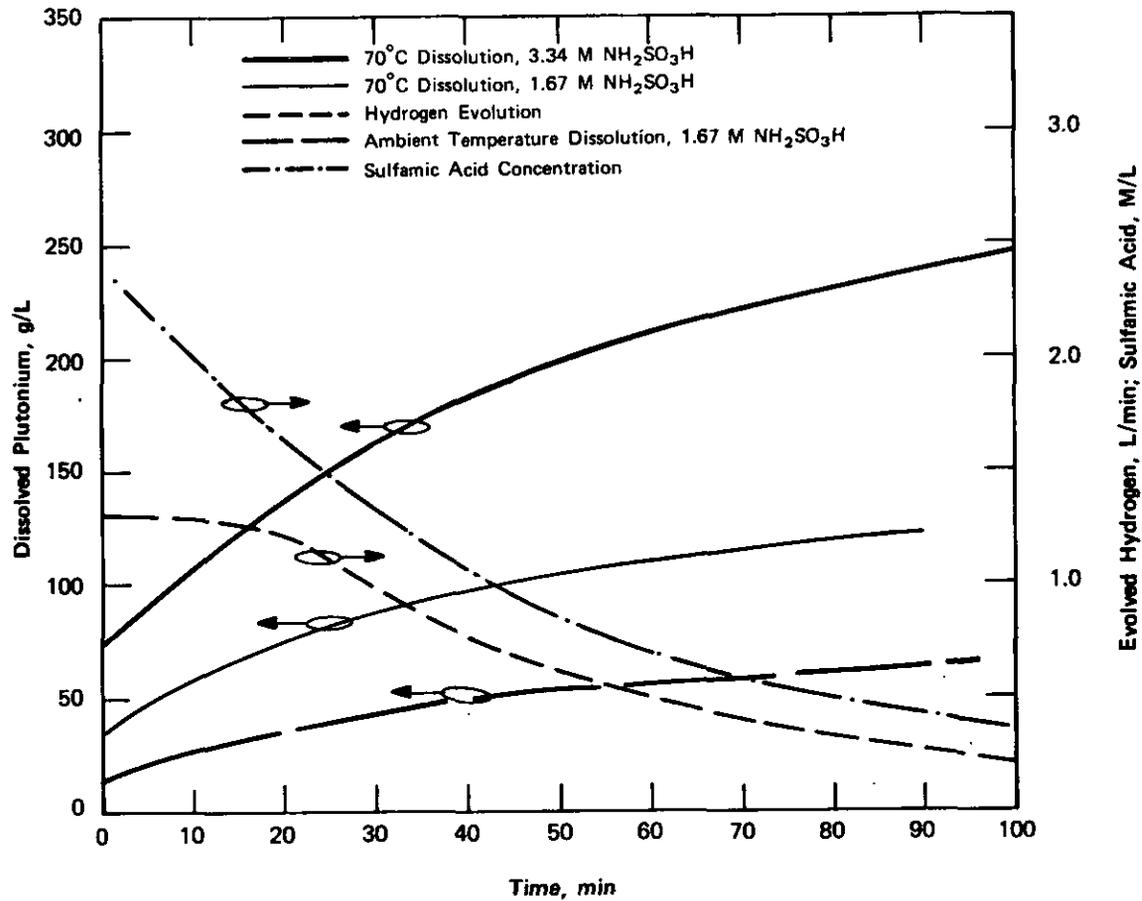


FIGURE 12. Batch Dissolution of Plutonium in 3.34M Sulfamic Acid (Case 6)

solution for dissolving from 125.2 cm² to 296.4 cm². For calculational purposes, this batch is assumed to begin with a residual plutonium concentration of about 75.18 g Pu/L, a residual sulfamic acid concentration of 2.34M, a button surface area of 296.4 cm², and a hydrogen off-gas rate of about 2.67 L/min. After a 0.25-hr dissolution interval, the plutonium concentration is calculated to have increased to about 168 g Pu/L and the hydrogen evolution rate would have decreased to about 2.0 L/min (Figure 13). At the end of a one-hr dissolution time, the plutonium concentration would be approaching 254 g Pu/L and the hydrogen off-gas rate would be about 1.0 L/min. This plutonium concentration compares to about 129 g Pu/L for a 70°C/1.67M sulfamic acid dissolution, and only about 60 g Pu/L for an ambient temperature/1.67M sulfamic acid dissolution. Continuation of dissolving for a total of 90 minutes would increase the plutonium concentration to about 262 g Pu/L.

Again for convenience of comparison, the calculated plutonium concentrations with time for 1.67M sulfamic acid both at ambient temperature and at 70°C are included in Figure 13.

APPLICATIONS TO RECOVERY OPERATIONS

Dissolution of plutonium metal on a production scale consists of the following steps:

- Charging metal to the dissolver.
- Charging sulfamic acid solution to the dissolver.
- Dissolving the plutonium metal for a period of time.
- Displacing two-thirds of the plutonium solution with fresh sulfamic acid solution.
- Filtering the displaced solution to remove entrained sludge.
- Transferring the filtered solution to an accountability tank.
- Diluting the filtered solution with 3M nitric acid and transferring it to storage tanks to await processing through the Purex process.

Recent studies have shown that it is desirable to remove the sulfamate ion by precipitation as sulfamic acid if ingrown ²⁴¹Am is to be recovered from the solvent extraction waste stream. This procedure consists of adding 2L of chilled 72% nitric acid per liter of plutonium sulfamate solution. Additional sulfamic acid can be precipitated and filtered by

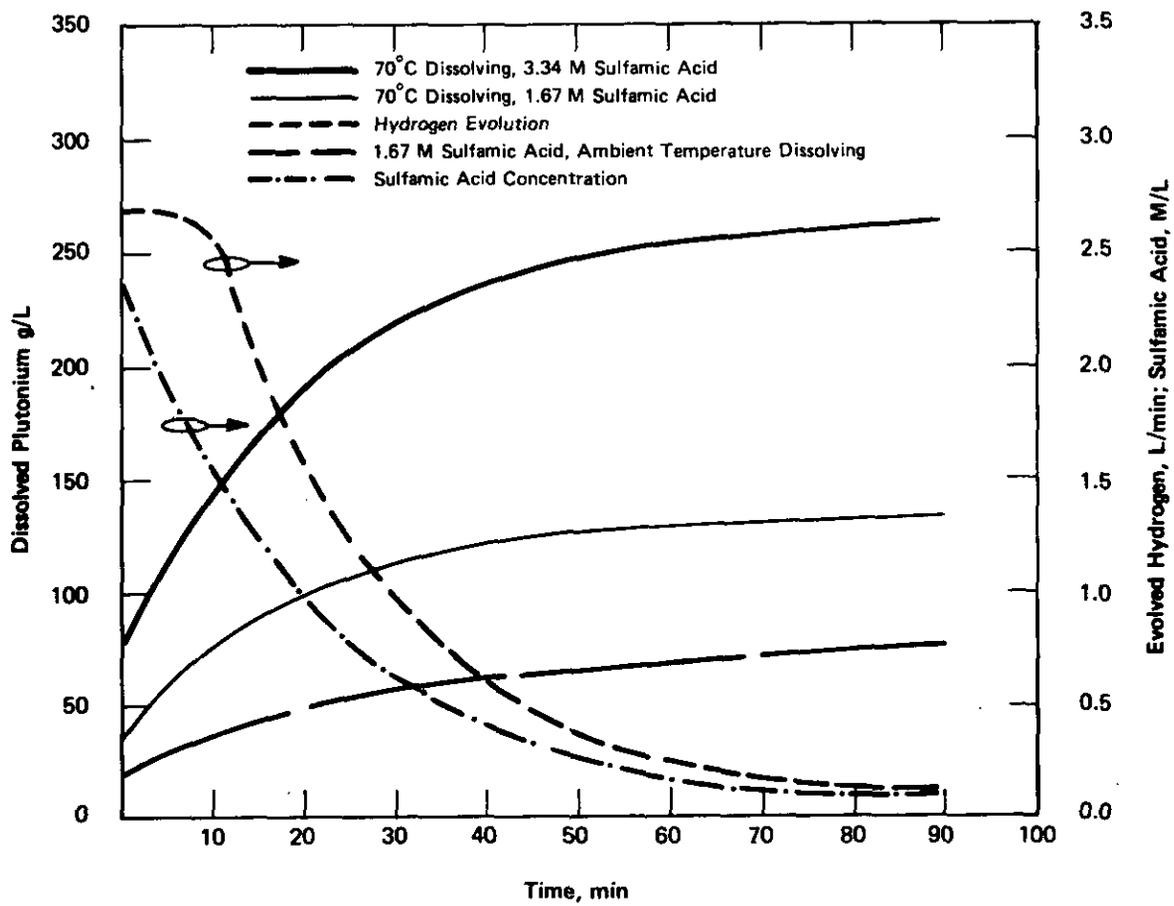


FIGURE 13. Batch Dissolution of Plutonium in 3.34M Sulfamic Acid (Case 7)

chilling the combined solutions to as low a temperature as practical, e.g. $\sim -10^{\circ}\text{C}$.⁴

These downstream operations require that the cycle time for dissolving plutonium be approximately 0.75 to 1.5 hr, depending upon whether or not the sulfamate ion is removed, the manner of diluting the solution, and other normal production problems. The advantages of the faster dissolving rate, of less sludge formation, and of a lower sulfamate-to-plutonium ratio can be obtained by dissolving plutonium at 65 to 80°C and more nearly exhausting the hydrogen ion. Raising the process temperature to this range will double the dissolving rate, double the plutonium concentration, and reduce the sulfamate-to-plutonium ratio from ~ 3.5 -to-1.

An even faster dissolving rate could be obtained by adding water and solid sulfamic acid to the dissolver. At 70°C, the solubility of sulfamic acid is twice its solubility at 25°C. Therefore, operation at 70°C in this mode would increase the overall throughput rate by a factor of about four over previous ambient temperature operations without changing the cycle times. Operation at elevated temperatures, however, would require a refrigeration unit to cool the solutions before nitric acid could be added for sulfamic acid removal.

Maximum benefit to downstream processing would occur if the dissolver were operated at 70°C with solid sulfamic acid added to the dissolver. This combination would allow the lowest sulfamate-to-plutonium ratio at the end of the precipitation of the sulfamic acid; this will decrease the sulfamate-to-plutonium ratio to less than 10% of the ratio of past campaigns.

The latter mode of operation would mean that plutonium liquors would leave the dissolver at approximately 200 to 250 g Pu/L and a sulfamate ion concentration of about 3.3M. After filtration, 2 L of solution would be chilled to ambient temperature or lower before adding 4 L of chilled 72% nitric acid (-10°C or lower) to the tank. After a 30-min wait time (refrigeration unit continuing to cool) for complete precipitation, the chilled solution would be filtered to remove the precipitated sulfamic acid. After filtration, the white sulfamic acid crystals would be washed with a volume of chilled 72% nitric acid. After changing valving to route the filtrate to waste, the sulfamic acid crystals would be washed with a small volume of cold water ($\sim 4^{\circ}\text{C}$) to remove residual nitric acid. The product can then be air dried and reused. Alternatively, the sulfamic acid crystals could be dissolved in water (ambient to boiling) and transferred directly to waste.

The plutonium nitrate solution, now containing about one-tenth of the original sulfamic acid, would be sampled, diluted

to ≤ 6 g/L with 3M nitric acid and transferred to canyon storage to await processing through the second plutonium cycle or for use as isotopic blend material for first-cycle processing.

In periods of metal dissolving when ^{241}Am recovery is not to be considered, maximum advantage of the faster dissolving rates at elevated temperatures could be gained. Charging water and solid sulfamic acid to the dissolver at 70 to 75°C would yield the faster rates. Cycle time could be shortened to the minimum time necessary for downstream filtration, sampling, and transfer. A dissolving rate of about 600 to 800 g/hr per dissolver could thus be achieved, with the transfer system being the limiting factor.

In addition to the faster dissolving rates, another advantage to overall throughput rate is the decreased amount of sludge formed. As a result, filtration times would be shorter, and sludge dissolving periods would be minimized. The possibility of fires involving the sludge would also be minimized.

An additional option becomes available if the sulfamate ion is removed. While the solution is in the high concentration of nitric acid ($\sim 10\text{M}$), the residual sulfamate could be destroyed by irradiation with UV light. Irradiation of nitric acid with UV light generates nitrous acid and oxygen. The nitrous acid would in turn react with the sulfamic acid to form nitrogen gas and sulfuric acid.

Elevated temperature dissolution and precipitation of the sulfamic acid followed by oxidation of the residual sulfamate with UV light will reduce the amount of sodium fed to the evaporators to only a few percent of that produced by the present process. The volume of solution could then be reduced sufficiently to perhaps allow direct precipitation of the ^{241}Am as oxalate, assuming other impurities (i.e., plutonium or the stainless steel corrosion products Fe, Cr, Ni) were sufficiently low.⁵ If these impurities were reduced or eliminated from the solution to allow direct precipitation of the ^{241}Am , the number of cation column runs required for further cleanup of the ^{241}Am would be only about 1 or 2 runs per kg ^{241}Am .

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