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A MONITOR FOR RADIOIODINE IN REACTOR COOLING WATER

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A MONITOR FOR RADIOIODINE IN REACTOR COOLING WATER

By

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November 5, 1964

Hanford Atomic Products Operation
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INTRODUCTION

The purpose of this study was to demonstrate the feasibility of a method for monitoring I^{131} in reactor cooling water. The presence of iodine isotopes in reactor cooling water was encountered as an interference in the development of an As^{76} monitor.[1] Work directed to removal of the interference suggested the possibility that a system could be developed to monitor radioiodine, specifically I^{131}.

This report describes the progress that has been made toward the development of an I^{131} monitor.

SUMMARY

A system was developed for monitoring radioiodine in reactor cooling water. Carbon tetrachloride is used to extract the iodine isotopes from the effluent sample stream. A gamma detector is used to measure gross gamma activity in the carbon tetrachloride.

The feasibility of this system was demonstrated. The I^{131} concentration in the effluent during normal reactor operating conditions is a reasonably constant fraction of the total iodine. Increased iodine activity was observed during rupture conditions.

DESCRIPTION

Monitor Design

The CCl_4 extraction technique for the analysis of iodine isotopes[2] was adapted to the monitor system. CCl_4 was mixed with a pre-treated reactor effluent stream and radioactivity from the extracted iodine isotopes in the CCl_4 phase was measured. Continuous flows of CCl_4 and the reactor effluent sample were maintained through the monitor. A simple gamma ray spectrometer with count rate meter-recorder output was used to measure the activity.

Sample System

The flow diagram of the monitor is shown in Figure 1. Reactor cooling water is fed to the extraction cell and pre-treated with $NaNO_2$ solution, (50 grams/liter of H_2O) and H_2SO_4-KI solution (15 ml conc. H_2SO_4 and 0.133g KI/liter of water). The KI carrier is oxidized to I_2 by the $NaNO_2$ in the acid condition. The solution is then stirred down into the CCl_4 layer with a hollow stem extraction stirrer; see Figure 2. The water overflows from the top of the cell to the drain. The CCl_4 drains from the bottom and overflows to the counting cell. Flow rates are indicated in Figure 1.

The sample cell, shown in Figure 3, holds 50 ml. The I_2 enriched CCl_4 enters at the bottom of the cell and overflows from the top. The CCl_4 overflow is fed to a glass bead packed column of 10 per cent $NaHSO_3$ solution. The $NaHSO_3$ reduces the I_2 to I^- which is stripped from the CCl_4 phase. CCl_4 overflow (taken from the bottom of the column) is fed to a glass bead packed wash column. Traces of I^- are washed from the CCl_4 by a counter current stream of distilled water. CCl_4 overflow (from the bottom of the column) goes to a storage tank for return to the system. CCl_4 is metered into the extraction cell from storage with a positive displacement type pump*. A peristaltic action pump** was used to control the water sample flow rate. Chemical addition was controlled by gravity flow through an orifice.

Detector System

Gamma activity in the CCl_4 is detected by a 3" NaI (Tl) crystal coupled to a 3" phototube. The detector is mounted directly under the counting cell. Background radioactivity is reduced by 4 inches of lead shielding. The phototube anode signal goes to a preamplifier, a linear amplifier and a discriminator set to accept gamma energies above 0.1 Mev. The discriminator output is fed to a count rate meter and strip chart recorder.

A block diagram of the electronics is shown in Figure 4.

DISCUSSION

A typical record of the monitor during a reactor shutdown is shown in Figure 5. The lag in activity level change is caused by slow response time of the monitor. The rise in activity following shutdown is probably caused by

* Mini-pump, Milton Roy
** Sigma motor pump

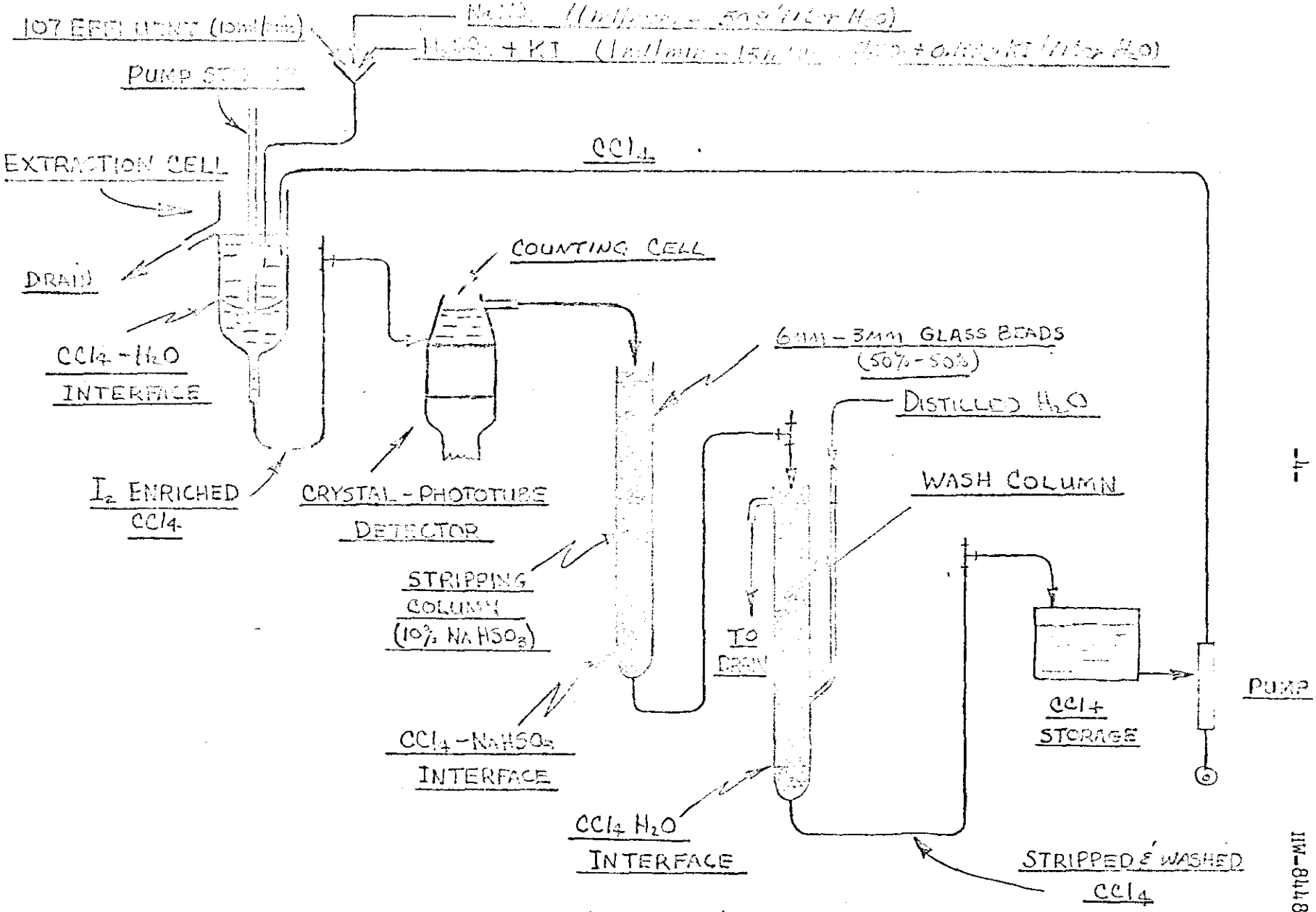


FIGURE 1

I_2 MONITOR - FLOW DIAGRAM

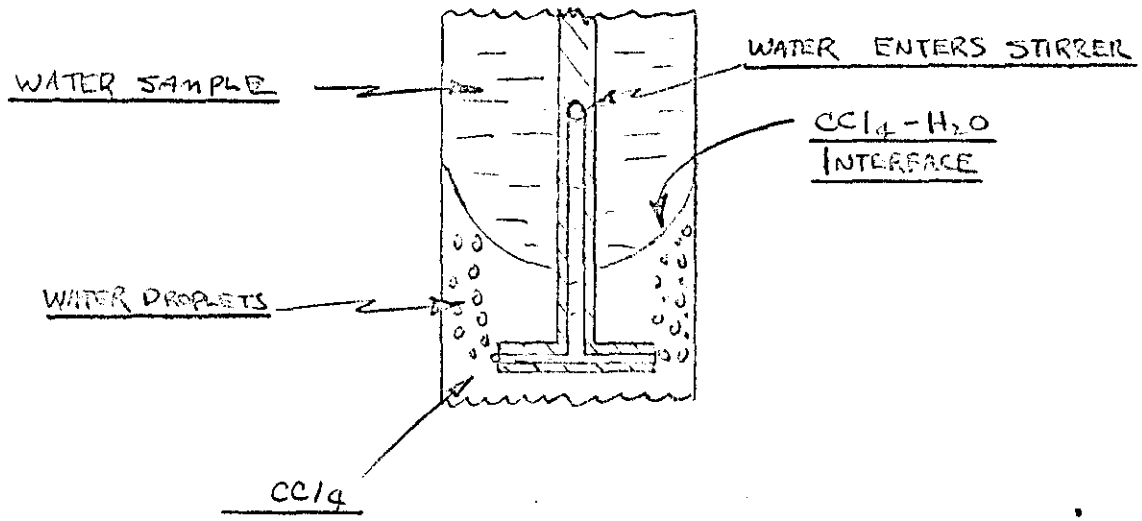


FIGURE 2. EXTRACTION STIRRER

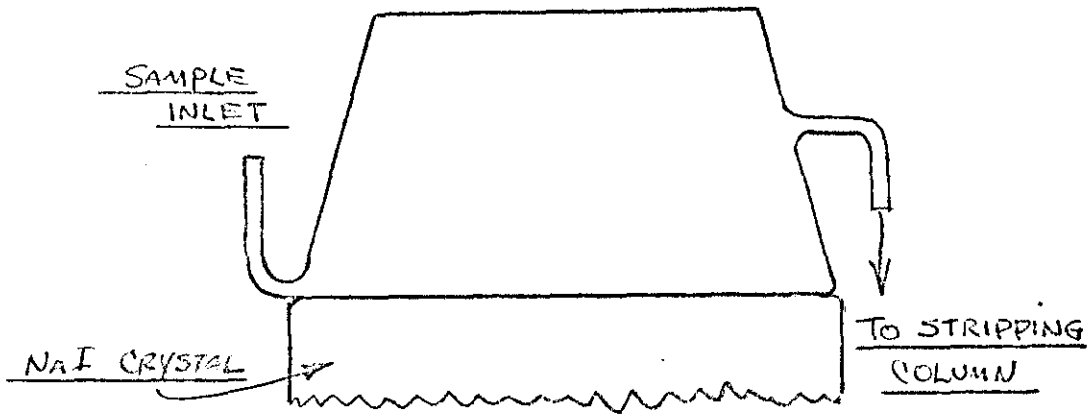


FIGURE 3. COUNTING CELL

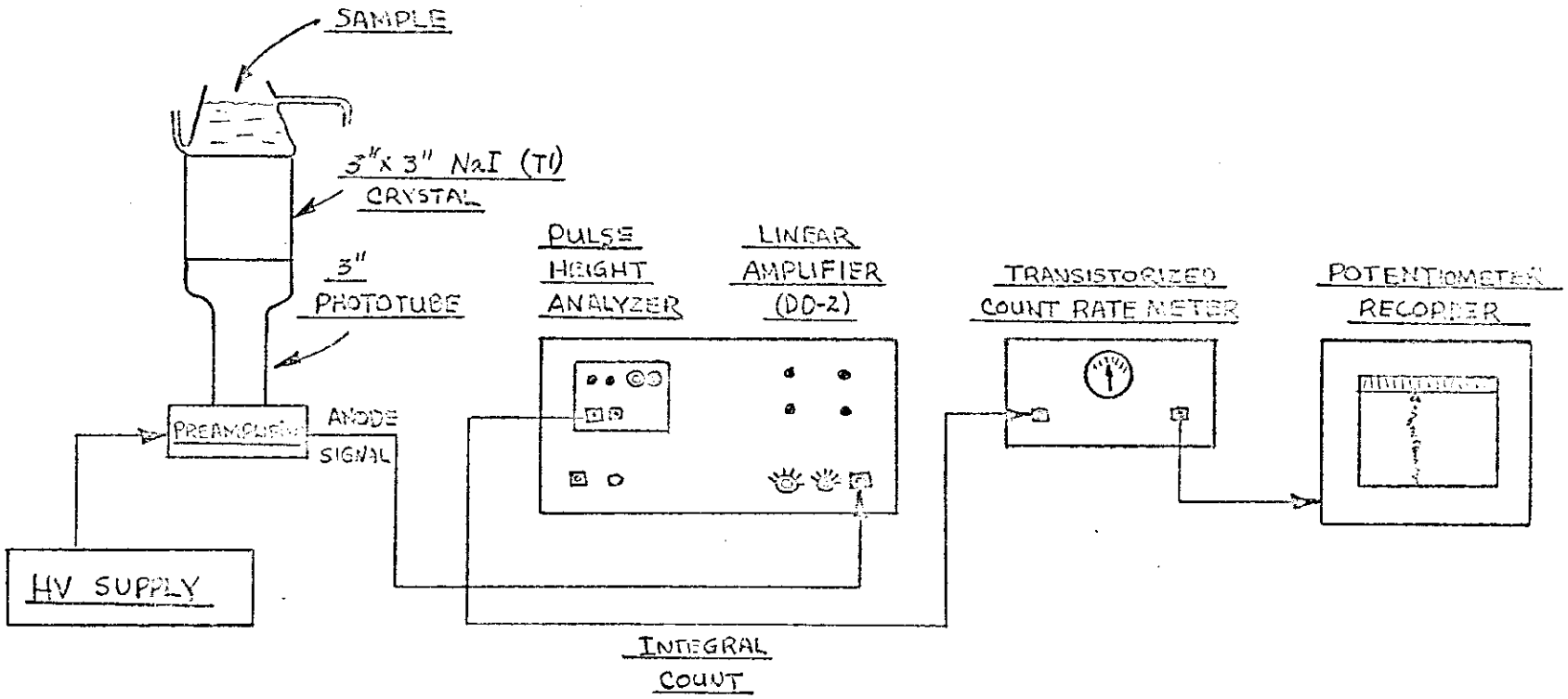
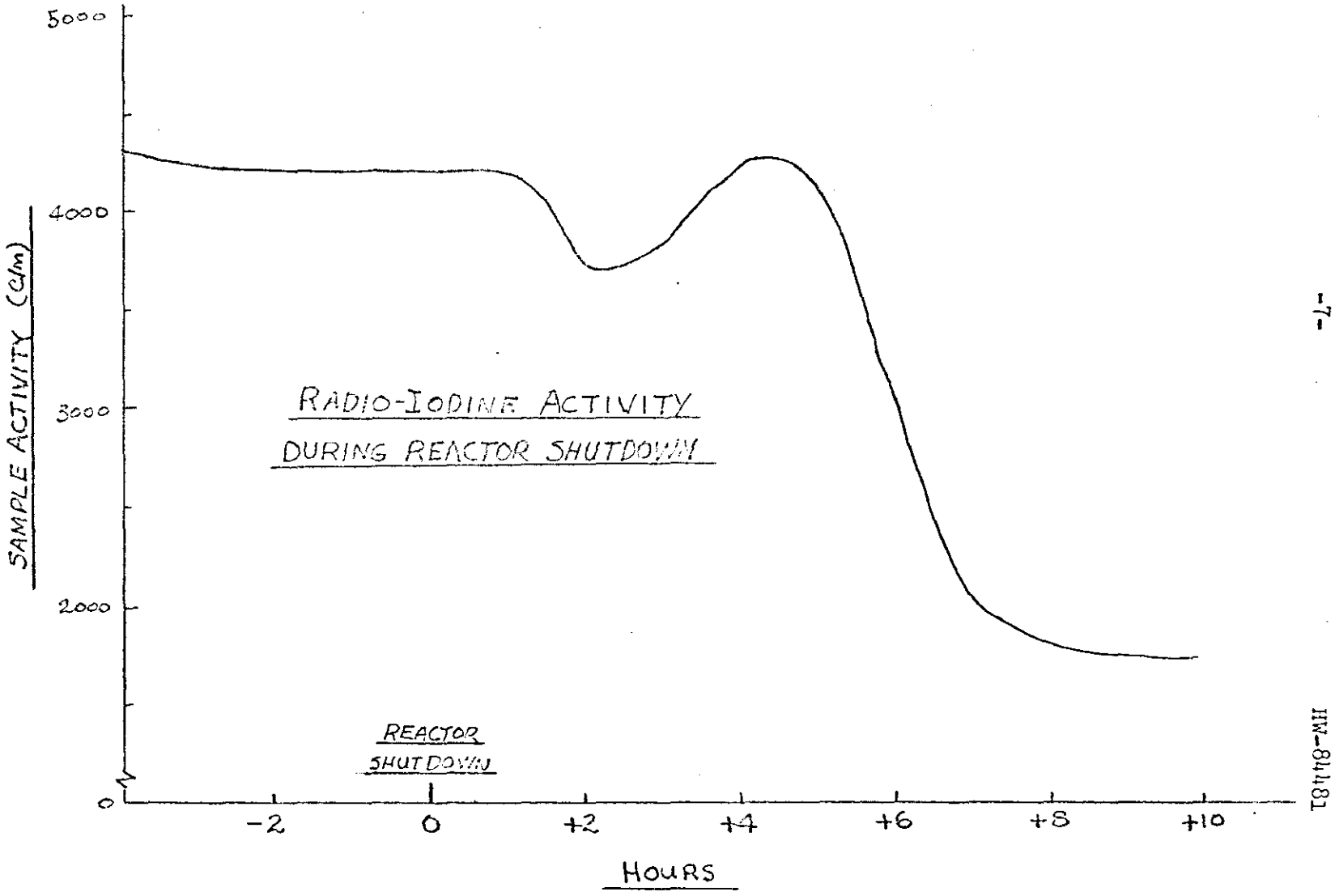


FIGURE 4. IODINE MONITOR COUNTING SYSTEM

FIGURE 5



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decreased water flow through the reactor. Activity buildup following reactor startup is shown in Figure 6.

Two ten-day periods of unattended operation demonstrated reliable performance. During one of the periods, a full element rupture occurred; the activity recorded by the monitor is shown in Figure 7. Reactor shutdown time nearly coincides with the off-scale activity indication because of slow response time of the monitor (30 minutes for first indication and 70 minutes for a 66 per cent change). Another rupture condition was monitored at increased flow rates with response time adjusted to 15 minutes for a 66 per cent change. The activity buildup is shown in Figure 8.

CALIBRATION

No direct calibration of the monitor was made, however, iodine isotope ratios were measured. I^{131} is the most important iodine isotope because of the 8.5 day half-life. If the ratio of I^{131} to other iodine isotopes is constant, the total iodine activity indicated by the monitor could be used to determine I^{131} concentrations. Accordingly, ratios of I^{131} vs. I^{132}, I^{133}, I^{134} and I^{135} were measured. Samples were collected at random time intervals from the monitor inlet. Analyses were made to determine I^{131}, I^{132}, I^{133}, I^{134}, and I^{135} concentrations in the sample at the time corresponding to measurement by the monitor. The ratios for 5 samples are shown in Table I.

TABLE I

I^{131} VS. IODINE ISOTOPE CONCENTRATIONS

Sample	I^{131}/I^{132}	I^{131}/I^{133}	I^{131}/I^{134}	I^{131}/I^{135}
1	0.0396	0.0551	0.00704	0.0200
2	0.0193	0.0509	0.00604	0.0242
3	0.0188	0.0615	0.00681	0.0160
4	0.0215	0.0452	0.00700	0.0233
5	0.0206	0.0466	0.0921	0.0206
Average	0.0220	0.0519	0.00726	0.0208
Standard Deviation	±41%	±15%	±16%	±15%

The high standard deviation in the case of I^{131}/I^{132} ratio cannot be explained; however, problems were encountered in sample analyses which may have contributed, in part, to the high value. The samples were collected during periods of normal reactor operation.

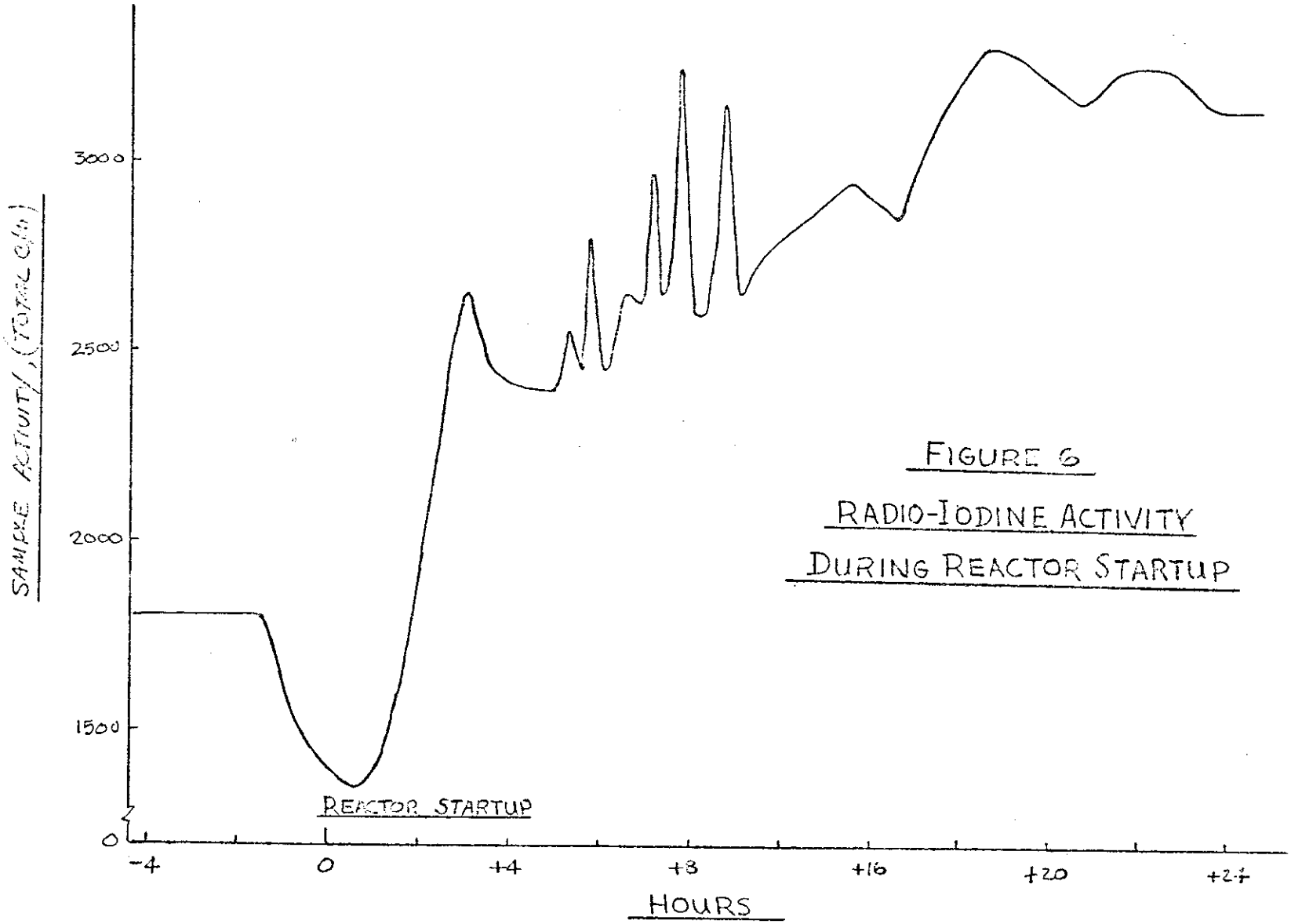

FIGURE 6

RADIO-IODINE ACTIVITY

DURING REACTOR STARTUP

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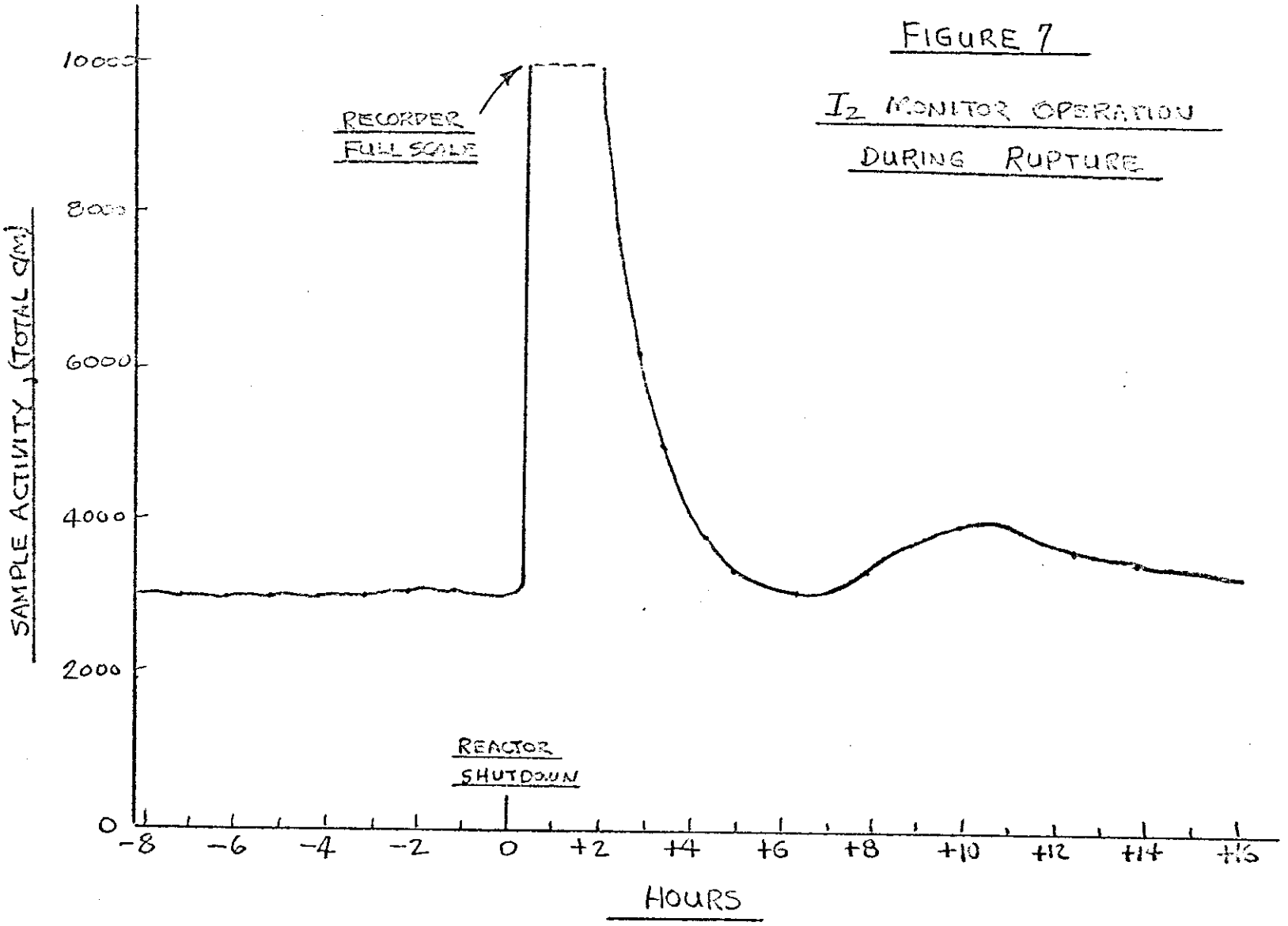

FIGURE 7

I_2 MONITOR OPERATION DURING RUPTURE

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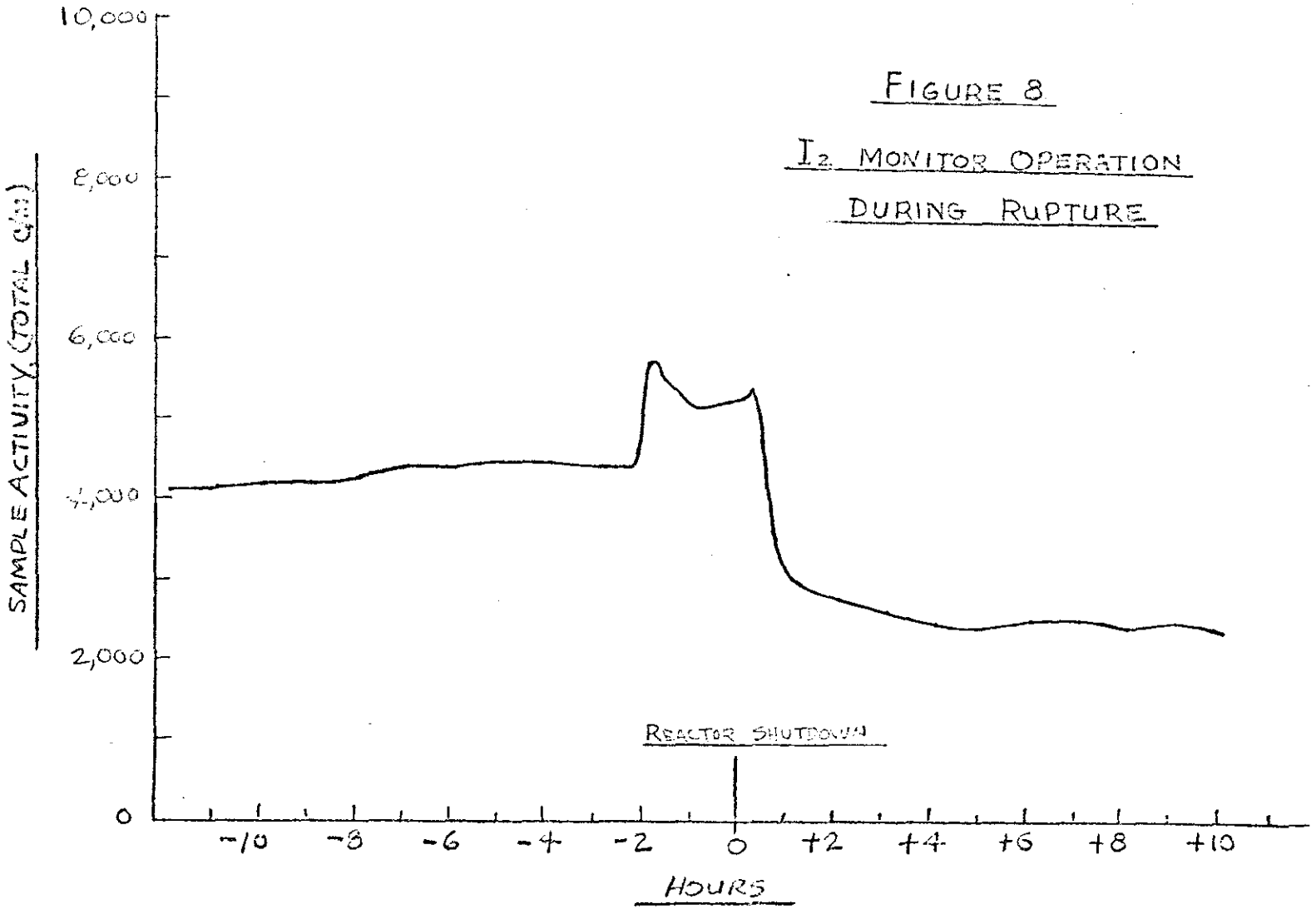

FIGURE 8

I_2 MONITOR OPERATION

DURING RUPTURE

No samples were collected during periods of abnormal reactor operation such as a rupture event. Samples of the CCl_4 in the counting cell were collected and checked for presence of interfering isotopes. None were found, except in cases where entrained water droplets were carried over to the counting cell by the CCl_4 stream.

CONCLUSIONS

Reliable operation of the monitor was obtained with a minimum of attention. The radioiodine separation appears clean with no interfering isotopes in the CCl_4 phase. The I^{131} concentration in the sample stream is a reasonably constant fraction of the total iodine.

The major problem encountered was handling the CCl_4. Several pumps were tried and all were unsatisfactory except a positive displacement piston type pump with a two stage ball check valve system. A constant head orifice was tried but plugging caused unsatisfactory operation. An occasional problem with entrained water droplets in the CCl_4 stream was encountered; however, this was remedied by changing the stirring pump-extraction cell configuration.

Further work is required to determine the I^{131}/I_2 ratio during abnormal reactor operation. Sample analyses are required to establish the variation of I^{131} concentration with total radioiodine activity.

The behavior of the monitor during a rupture condition would indicate possibility of use as a rupture detection system. The minimum response time is limited to approximately 15 minutes and therefore the time element is an unattractive feature.

The recommendations for further work are to:

1. Investigate methods of selective gamma energy analysis to determine I^{131}; possibly from I^{134} - I^{135} measurements.
2. Determine the I^{131}/I_2 ratio during rupture conditions and at reduced and changing power levels.
3. Develop system for reducing or subtracting background.
4. Investigate use of bulk CCl_4 with one pass through the system and evaluate resulting hazards.

REFERENCES

[1] H.G. Rieck. An Automatic Monitor for As^{76} Concentrations in Reactor Cooling Water, HW-75374, April 1963.

[2] J. Kleinberg, G.A. Cowan. The Radiochemistry of Fluorine, Chlorine, Bromine and Iodine, NAS-NS-3005, January 1960.